REPORT TO SPONSOR:

**RADIATION LIMITS FOR Nb_3Sn SUPERCONDUCTORS
FOR ITER MAGNETS:
A LITERATURE REVIEW**

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EXECUTIVE SUMMARY

Results of a literature search on the radiation sensitivity of Nb₃Sn conductors are reported. An introductory section examines differences in newer Nb₃Sn conductor fabrication techniques, since most data were acquired on older, bronze processed material. Then, data on the radiation sensitivity of the transition temperature, the critical temperature, the critical current, and the upper critical field are compiled. This data base is assessed to determine radiation limits for the use of these conductors in ITER TF (toroidal field) magnets.

In contrast to the limited irradiation data base on ITER magnet insulation, the Nb₃Sn data base contains extensive studies by investigators from different institutions of the radiation sensitivity of relevant 4-K performance parameters. Since limited recovery (about 25%) occurs at 300 K and full recovery does not take place until about 800°C, ambient-temperature irradiation results are often similar to cryogenic irradiation results, and provide important substantiation of the more limited cryogenic data base. However, the high fluence, fast neutron ($E > 0.1$ MeV) cryogenic irradiation data base is extremely limited for ternary (or alloyed) Nb₃Sn, and ternary material is significantly more sensitive to radiation than binary material. Part of the ternary high fluence data base is derived from 14-MeV irradiation, and conversion to an ITER TF magnet fast neutron fluence ($E > 0.1$ MeV) is problematical. Several experimental tests of the conversion factors have given unsatisfactory results, especially for the critical current, J_c . Furthermore, the ternary Nb₃Sn prototype specimens tested at 14 MeV just before cryogenic testing was discontinued had J_{cs} at 15 T from about 370 to 450 A/mm², which might not meet present ITER requirements. Current ITER procurement contracts specify J_c values of 550 and 700 A/mm² at 12 T. A higher-performance ternary material would probably be more radiation-sensitive, especially if J_c were increased by increasing ρ_n , the normal-field resistivity. Also, several investigations indicate that the J_c at 12 T would be more degraded by radiation than the J_c measured at 15 T.

The available cryogenic fast neutron data, obtained on an (Nb,1.5 wt.% Ti)₃Sn specimen at 8 T and 5 K, suggest an ITER radiation limit of $2.5 \times 10^{21}/\text{m}^2$ ($E >$

0.1 MeV). The J_c of this modified jelly roll material at 12 T was probably $\sim 1000 \text{ A/mm}^2$, but was not specifically reported. Conversion of the cryogenic 14-MeV data on an $(\text{Nb}, 1.1 \text{ wt.}\% \text{ Ti})_3\text{Sn}$ specimen ($J_c \approx 450 \text{ A/mm}^2$) allows a limit equivalent to $\sim 6 \times 10^{22}/\text{m}^2$ ($E > 0.11 \text{ MeV}$, fission spectrum). Some authors recommend against conversion of 14-MeV J_c fluences to fission fluences, because the 14-MeV neutron damage cascades may allow more recombination of defects than the fast neutron damage cascades.

A conservative assessment of the data base suggests a radiation limit for ternary Nb_3Sn conductors of $2.5 \times 10^{21}/\text{m}^2$ ($E > 0.1 \text{ MeV}$, magnet spectrum). (The ITER TF magnet fluence should be set at about 75% of the limiting fission fluence of $3.3 \times 10^{21}/\text{m}^2$, owing to a harder spectrum.) Since this radiation limit is based upon cryogenic fast neutron irradiation of only one specimen type, further testing of ITER prototype conductors is strongly recommended. Additional comparisons with experiment of expressions for the combined irradiation and strain dependence of superconducting transition temperature, critical current, and upper critical field are also desirable.

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1. INTRODUCTION

The data base on radiation damage to Nb₃Sn superconductors is compiled from the literature and assessed in this report. Nb₃Sn superconductors are currently under procurement for use in ITER magnet prototypes. In contrast to the data base on insulation materials proposed for use in ITER magnets, the data base on the radiation damage of Nb₃Sn is much more complete. Key results have often been confirmed by several groups at different institutions. The investigation of variables that influence radiation damage has also been much more complete for Nb₃Sn than for insulators. Furthermore, in situ testing of superconducting parameters is much easier than in situ mechanical testing of insulators, and in situ testing has invariably been performed after cryogenic irradiation of Nb₃Sn. However, in recent years, Nb₃Sn testing has also suffered from the lack of 4-K irradiation facilities. Just as new processing methods to obtain more economical Nb₃Sn conductor products in large quantity were being developed, cryogenic irradiation sources were being phased out. Therefore, this brief introductory section presents some basic information on the properties and structure of Nb₃Sn superconducting composites and the distinctions between different fabrication processes. This provides a background to assess the adequacy of the current cryogenic data base on radiation damage. Also, since synergistic effects of strain and irradiation have recently been investigated, a brief discussion of the effects of strain on Nb₃Sn properties is included in this introduction. Söll [1993] has recently reviewed the state of the Nb₃Sn data base.

1.1. Nb₃Sn SUPERCONDUCTING PROPERTIES

The critical temperature, T_c , and the upper critical field, B_{c2} , are intrinsic properties of the A15 superconductor Nb₃Sn that are determined by the composition and the structure. Typical values are 18 K for T_c and 20 T for B_{c2} at 4 K [Turck, 1991]. B_{c2} is related to T_c by

$$B_{c2} \approx \rho_n \gamma T_c, \quad (1.1.1)$$

where ρ_n is the electrical resistivity of the material in the normal state, and γ is the coefficient of the electronic contribution to the heat capacity. (This expression is valid in the "dirty limit" of the Abrikosov theory where the electronic mean free path in the normal state is much less than the

coherence length at 0 K.) Increasing T_c and B_{c2} implies some change in the chemistry of the material by either modifying the proportions or by introducing other elements. It is not surprising, therefore, that T_c and B_{c2} are (usually) not significantly improved by radiation, but can only be degraded. (However, if ρ_n increases with radiation damage faster than T_c decreases, an augmentation of a few degrees can be observed [§§3.2.3,3.3].) The critical current density, J_c , however, is not an intrinsic property, but can be improved by modifications of the microstructure provided during the final metallurgical processes, such as cold work and heat treatment. J_c is the result of the pinning of the lines of magnetic flux that penetrate the material in the mixed state. The capability of various imperfections such as defect cascades, impurities, dislocations, and voids to inhibit the fluxoid motion leads to an increase in J_c . (See Brown et al. [1975] for further discussion.) Since J_c can be increased by increasing the pinning force, that is, by optimizing the sizes and separations of the pinning sites, either enhancement or degradation could occur from radiation damage. The pinning mechanism generally is dominated by grain boundaries and grain size in binary, unirradiated Nb_3Sn [Turck, 1991]. The Nb_3Sn phase is formed by heat treatment at the end of wire manufacture. The heat treatment conditions determine the grain size and grain boundary properties, and thus, the pinning behavior. The pinning force varies inversely as the grain size. Therefore, the duration and temperature of the heat treatment is a compromise between Nb_3Sn layer growth that increases the superconducting volume fraction (and thus, I_c) and the grain growth that decreases the pinning force (and thus, J_c). Hence, it is not surprising that both ternary alloying and radiation damage, by supplying further pinning sites, can improve J_c above the values obtained through metallurgical processing.

1.2. Nb_3Sn SUPERCONDUCTING COMPOSITE STRUCTURE

To keep self inductances and operating voltages at acceptable levels, a monolithic Nb_3Sn conductor is not a viable option. Also, a thick conductor is not amenable to the cold-working that is required. Therefore, for a large I_c , a cable must be assembled from superconducting wires. Low ac losses can be obtained by a decrease in the dimensions of filaments, strand, and the cable, and of the stabilizer fraction. The Cu fraction is chiefly determined by the

requirements of thermal stability. The superconducting filaments are distributed in a continuous matrix of resistive material (cupronickel or bronze) and the stabilizing Cu is located either in the middle or in an outer shell. Since the highest losses due to coupling currents between filaments occur when the Cu is located around the axis of the wire, it has been found desirable to locate the Cu at the periphery and to reduce the radius of the filamentary bundle as much as possible [Turck, 1991]. However, some Cu at the axis improves thermal stability with respect to small perturbations, and drawability and fabricability are also improved. For thick wires, filaments are distributed in bundles of about 0.2 to 0.5 mm in diameter to provide Cu paths for heat to be removed and to limit the size of low conductivity regions. Barriers inside the Cu matrix may also decrease ac losses.

1.3. Nb₃Sn SUPERCONDUCTOR FABRICATION

Owing to the considerations discussed above, and others not detailed here, several methods of Nb₃Sn conductor fabrication are presently used to optimize properties. Because of the brittleness of the Nb₃Sn phase, in all methods only the ductile Cu, bronze, or Nb is drawn, and the Al₅ phase is formed at the end of the process. Three fabrication processes used in current ITER procurement contracts will be briefly described. Recent ITER procurement specifications for superconductors are given in Table 1.3.1, from Minervini [1994]. The following discussion of fabrication processes is based largely on reviews by Turck [1991] and Thöner et al. [1991]..

1.3.1. Bronze Process

The bronze process has been in use since the early 1970s; hence, much of the Nb₃Sn irradiation data pertains to bronze processed specimens. Magnet wire for MRI (magnetic resonance imaging) applications is usually made by the bronze process, which has been the most common way to produce large amounts of conductor. The wires have a regular arrangement of filaments that are 2 to 5 μ m in diameter, ac losses are low, and performance is predictable. The production billets contain Nb in CuSn bronze rods. The stabilizing Cu is protected from Sn diffusion by an Nb or Ta barrier (Ta is preferred to avoid formation of the Al₅ Nb₃Sn phase in the diffusion barrier, with resulting

Table 1.3.1. Specifications and Procurement Program for ITER Conductor, March, 1994. From Minervini [1994].

Strand Grades Specifications for Critical Current and Hysteresis Loss

Grade	$J_c(12\text{ T}, 4.2\text{ K}, 0.1\text{ }\mu\text{V/cm}, \epsilon_{\text{applied}}=0)$ based on Non-Cu Volume (A/mm^2)	Hysteresis Loss for $\pm 3\text{ T}$ cycle based on Non-Cu volume (mJ/cm^3)
High Performance 1 (HP-1)	≥ 700	≤ 600
High Performance 2 (HP-2)	≥ 550	≤ 200
Low Performance 1 (LP-1)	≥ 550	≤ 600

Staged Procurement Program (continued)

- The following contracts are in place in the U.S. program

Contracts with MIT:

- ☐ TWCA - 3.75 metric tons of strand by MJR process to meet ITER HP-1 spec. Stages 1 and 2 released
- ☐ IGC - 3.75 metric tons of strand by internal tin process to meet ITER HP-1 spec. Stages 1 and 2 released
- ☐ Supercon - 70 kg of bronze process strand to meet ITER HP-2 spec.

Contracts with LLNL:

- ☐ Oxford - 250 kg of strand by internal tin process to meet ITER HP-1 spec.
- ☐ IGC - 250 kg of strand by internal tin process to meet ITER HP-1 spec.

brittleness and possible ac losses, and possible Sn in the pure Cu. The same problem occurs in the internal Sn method.) The superconducting layer is formed by heat treatment at $\sim 700^{\circ}\text{C}$ for ~ 100 h. There are several limitations inherent in the bronze process. To avoid filament contacts after reaction, the distance between the rods that contain Nb has to be carefully determined and controlled. The bronze must be fabricated with a high degree of control of quality and composition. Since the maximum Sn content in the bronze is 13.5 wt.% (7.8 at.%), the amount of Nb_3Sn that can be formed, and thus, the I_c , is limited. Several partial annealings are required during the drawing process, making it more time consuming and costly. Despite the limitations noted above, the final product can be of good quality. Thöner et al. [1991] presented data detailing the advantages of bronze processed Nb_3Sn in several respects.

1.3.2. Internal Sn Process

To overcome some of the bronze process limitations and to reduce costs, intermediate annealing is eliminated in the internal Sn method by drawing Cu, Nb, and Sn. The method developed in Japan is known as the tube method. The method developed in the US by IGC involves a co-reduction of Cu, Nb, and Sn (or, Sn-rich) elements to final size. A composite tube of Nb filaments and Cu made by hot extrusion is filled by an Sn core, then drawn and restacked. During a low temperature heat treatment (200 to 400°C), the Sn diffuses into the Cu to form a bronze, in order to obtain a homogenization in the composition before the high temperature heat treatment to form Nb_3Sn . The Nb_3Sn filaments in the low Sn bronze matrix are surrounded by Ta diffusion barriers that protect the Cu stabilizer. The method has intrinsic advantages in producing fine filaments with high J_c . At the time of Turck's review (1991), problems existed regarding insufficient bonding between Cu shells of the bundles during cold drawing that limited the wire diameter to 1 mm. The Sn core could lead to irregularities and sausaging of strand structure along the length or even to ruptures during drawing. (A Cu matrix is much softer than a bronze matrix.) The filaments were usually much closer at the end and more flattened and irregular than in the bronze process, which could lead to bridging after the heat treatment.

1.3.3. Modified Jelly Roll Process

This process from Teledyne Wah Chang completely eliminates any hot extrusion and intermediate annealings. The filamentary structure is obtained from an Nb sheet that has to be slit and somewhat expanded before being rolled together with a Cu sheet and an Sn core. The desired number of hexagonal rolled rods are stacked in a billet that is drawn down to a wire. Although the filaments are actually interconnected from place to place, short circuiting occurs periodically at distances considerably over 100 m, well above the twist pitch length of the strand. This should not be detrimental to current sharing and ac losses. A typical heat treatment schedule includes three treatments around 250 and 350°C to form the bronze homogeneously and one treatment at about 650°C to form the Nb₃Sn phase. The resulting bundles are fairly irregular and the filaments are far from being circular, but the method is very cost effective.

1.3.4. Issues in Conductor Development

Especially in the internal Sn process, overall J_c and ac losses must be traded off. Bridging can be reduced by increasing the Cu content between unreacted Nb rods or meshes, but this reduces the superconducting volume fraction and therefore, I_c . Fine filaments ($<3 \mu\text{m}$) are more easily reacted fully, but thicker filaments are probably less sensitive to residual compressive strain and may enhance B_{c2} . Since an Sn concentration gradient occurs between the interfaces of the reacting Nb and the still unreacted Nb, T_c may vary over the reacted layer depth. This effect also reduces the allowable field and leads to scatter in the measurements of T_c , B_{c2} , and J_c . Alloying with Ti or Ta speeds up diffusion, improves J_c owing to extra pinning from precipitations and enhances B_{c2} through the increase of ρ_n [Equation (1.1.1)]. This allows a competitive improvement in J_c for Nb₃Sn produced by the bronze process. One example is the comprehensive investigation by Hampshire et al. [1985] of J_c , B_{c2} , and flux pinning for bronze processed (Nb,Ta)₃Sn. Additions of Ti generally decrease the workability. Large scale production costs are another significant issue, and may push development of new processing techniques.

Although the development of Nb₃Sn conductors was fairly mature by the time (~1990) that low temperature radiation testing virtually ceased, it is possible that the excellent data base obtained on principally bronze processed material may become outmoded and partly irrelevant to ITER needs, especially if high magnet fast neutron fluences (10^{22} to $10^{23}/\text{m}^2$) were reinstated or if processes other than the bronze technique were used for most of the conductor. Furthermore, most of the data base was obtained on binary Nb₃Sn conductors; ternary or alloyed conductors now appear necessary to meet the J_c requirements (Table 1.3.1), and the smaller data base on these materials indicates greater sensitivity to radiation, in general (§§2.3 and 3.3).

1.4. EFFECT OF STRAIN

Superconducting Nb₃Sn is sensitive to strain that is either longitudinal or transverse to a strand. J_c , T_c , and B_{c2} are reduced with strain. The effect of longitudinal strain is well established experimentally. Typical data for the critical current are shown in Figure 1.4.1 [Ekin, 1980]. The maximum in I_c or J_c occurs at a strain ϵ_m . This maximum arises from the longitudinal compressive prestress which the bronze matrix exerts on the Nb₃Sn reaction layer because of the difference in thermal contraction between the two materials as the composite is cooled from the reaction temperature of about 1000 K to the operating temperature near 4 K. The maximum occurs when the Nb₃Sn component experiences the smallest magnitude of intrinsic strain. An intrinsic strain, ϵ_0 , is defined as

$$\epsilon_0 = \epsilon - \epsilon_m, \quad (1.4.1)$$

where the negative values of ϵ_0 indicate the approximate longitudinal compressive strain and the positive values the longitudinal tensile strain. A J_c vs. strain diagram parameterized in this way is presented in Figure 1.4.2. This parameterization has been found to remove the discrepancies between different experiments. A typical longitudinal compressive prestrain from differential thermal contraction is about -0.3%. A stainless steel jacket around the superconducting cable increases the longitudinal compression to about -0.75% if applied in a wind and react technique and to about -0.4% if applied in a react and wind technique [citations, Mitchell and Bottura, 1991]. (With the wind and react technique, the jacket contributes to the thermal contraction from the reaction temperature on down to 4 K, whereas in the react

Figure 1.4.1. I_c of a specimen vs. uniaxial strain for fields ranging from 7 to 19 T. The strain ϵ_m is 0.32% for this specimen. The strain ϵ_{irrev} is about 0.8% for this specimen. From Ekin [1980].

Figure 1.4.2. Normalized J_c vs. the strain $\epsilon_0 = \epsilon - \epsilon_m$ for an Nb_3Sn specimen for fields ranging from 4 to 16 T. From Mitchell and Bottura [1991].

Figure 1.4.3. J_c vs. axial strain for Nb_3Sn wire. 1—Single wire. 2,3—Expected operating points for cable in conduit with stainless steel and Incoloy jackets. 2'—Expected operating point after electromagnetic loading. From Turck [1991].

and wind technique, the jacket is applied at ambient temperature after reaction.) Use of an Incoloy alloy with a thermal contraction that matches that of Nb_3Sn can eliminate some of the prestrain, but Incoloy may be more reactive than stainless steel with the components of the magnet insulation system at the reaction temperature.

Under electromagnetic loading, the compressive stress is partly compensated. Figure 1.4.3 shows the expected effects of axial strain on J_c for both Incoloy and stainless steel jackets and after electromagnetic loading [Turck, 1991]. Bending stresses also affect the final strain, as do stresses from various other fabrication techniques.

Transverse stresses affect J_c more than axial stresses do, and the effect is more significant in high magnetic fields, but fewer definitive experiments have been done. Figure 1.4.4 from Turck illustrates data from two investigations of the effects of axial and transverse strain on B_{c2} . Difficulties in assessing the transverse stress precisely may account for the differences in the results of the two investigations. The Young's modulus of the materials was about 100 GPa.

Synergistic effects of both radiation and strain have been little investigated. Sections 2.5 and 3.5 discuss the data found in this literature search. Summers et al. [1991] have presented equations for Nb_3Sn superconducting properties that include both strain and radiation damage (§§2.5, 3.5, and 4.3), but comparisons with experimental data have not been published at this writing.

Figure 1.4.4. Comparison of axial and transverse strain effects on B_{c2} of an Nb_3Sn wire from sources (1) and (2). Normalized to maximum B_{c2} . From Turck [1991].

2. RADIATION-INDUCED CHANGES IN THE TRANSITION TEMPERATURE

2.1. BULK Nb₃Sn

2.1.1. Ambient-Temperature Irradiation

Apparently, Swartz et al. [1964] first investigated the change with irradiation of the superconducting transition temperature, T_c , in Nb₃Sn. Using particle specimens irradiated to a fast neutron fluence of $1.5 \times 10^{22}/\text{m}^2$ ($E \approx 0.1$ to 4 MeV), they observed a decrease in T_c of 0.1 K. Bett [1974] irradiated commercial and laboratory specimens in the PLUTO reactor at AERE, Harwell to fast neutron fluences up to about $5 \times 10^{23}/\text{m}^2$ ($E > 0.7$ MeV) at about 70°C. The specimens were produced by coating Nb tape with Sn and then reacting at about 980°C. Some specimens were cleaned of unreacted Sn, and some were then annealed at 1150°C. One of the tape specimens had a Cu tape soldered to it. A similar depression of T_c with fluence was found for all batches of the material. The dependence of T_c upon fluence is shown in Figure 2.1.1 for one batch type. By extrapolation, the T_c of the non-preannealed material would decrease from 18 to 4 K after a fast neutron fluence of about $3.5 \times 10^{23}/\text{m}^2$. The figure also indicates that post-irradiation annealing temperatures of ~750 to 900°C were required to restore T_c to a temperature near to its original value. (The first onset of superconductivity was recorded as T_c in these measurements. In other investigations, T_c was sometimes measured in a different way, but this should not be a significant source of discrepancy, since ΔT_c vs. fluence is the desired result.)

Bauer et al. [1975] irradiated wire produced by an Sn diffusion process at 920 to 960°C in the BNL (Brookhaven National Laboratory) high flux beam reactor (HFBR) at temperatures expected to be near 80°C. Figure 2.1.2 shows a decrease in T_c to about 13.6 K (first onset measurement) after the highest fast neutron fluence of $5 \times 10^{22}/\text{m}^2$ ($E > 0.82$ MeV). Although the specimen preparation, irradiation conditions and fluence energy level were comparable to those of Bett, Bauer et al. found a much more pronounced decline in T_c with fluence, since Figure 2.1.1 shows a decline of T_c to only about 16.5 K at this fluence.

Figure 2.1.1 T_c vs. fast neutron fluence and its recovery after 2-h anneals at the indicated temperatures. From Bett [1974].

Figure 2.1.2. T_c change after fast neutron irradiation of Nb₃Sn diffusion wires at fluences of 3×10^{21} to $5 \times 10^{22}/\text{m}^2$. From Bauer et al. [1975].

Figure 2.1.3. Superconducting transition curves of Nb_3Sn for various fluences. From Sweedler et al. [1974].

An extensive investigation using the same HFBR facility also found that a fast neutron fluence of only $5.0 \times 10^{22}/\text{m}^2$ ($E > 1$ MeV) was required to lower the T_c to about 13.5 K [Sweedler et al., 1974, 1975]. These results on vapor-deposited Nb_3Sn specimens are shown in Figure 2.1.3. At a higher fluence of $5 \times 10^{23}/\text{m}^2$, T_c was reduced to about 2.8 K. Allowing for uncertainties in fluence measurement and effects of different specimen types, both these results and those of Bett indicated a serious level of damage, i.e., a drop in T_c of about 50%, at fast neutron fluences of about 1 to $2 \times 10^{23}/\text{m}^2$ (with $E > \sim 0.8$ to 1 MeV). Some corrections (increases) were applied later to the fluences reported in earlier papers [Sweedler et al., 1978], but these do not affect the numbers stated above because the authors had chosen the upper limit of the expected uncertainty in fluence in constructing Figure 2.1.3. A graph showing the depression in T_c for a number of A-15 compounds, with corrected fluence, is presented here as Figure 2.1.4. To convert the fast neutron fluence given in the figure for $E > 1$ MeV to a fast neutron fluence for $E > 0.1$ MeV, multiply by 4. This conversion gives the fast neutron fluence for a reactor spectrum, which can be approximately converted to an equivalent fast neutron fluence at the toroidal field (TF) magnet for a reactor of the ITER type by multiplication by 0.75. Thus, significant degradation in T_c ($\sim 50\%$ decrease) would occur at an ITER fast neutron fluence of about $3 \times 10^{23}/\text{m}^2$ ($E > 0.1$ MeV), according to the data of Figure 2.1.4. At an ITER fluence of $10^{23}/\text{m}^2$, a decrease of about 20% would be expected. For an ITER TF magnet fluence of $10^{22}/\text{m}^2$, only a very small change in T_c would be expected, based upon these ambient-temperature results on bulk Nb_3Sn .

Karkin et al. [1976] also carried out ambient-temperature neutron irradiation that verified the results of earlier investigations on T_c , as indicated in Figure 2.1.5. Details about the irradiation energy spectrum were not provided. Bulk specimens were prepared by baking tablets of pressed Nb and Sn powders for 20 h at 1050°C. Repeated powdering, pressing, and annealing under dynamic vacuum was carried out until the characteristic X-ray pattern of Nb_3Sn was obtained.

connected with compressive strains should be invoked for the faster decrease of T_c with dose of the alloyed material.

Weiss et al. advanced the following explanation for the data in Figures 3.3.3 and 3.3.4. Following Equation (3.2.2), the increase of B_{c2} in the low fluence region ($<10^{22}/\text{m}^2$) was attributed to an increase in the normal state resistivity ρ_n . In this fluence region, T_c and γ , the electronic density of states, do not change appreciably. The increase in B_{c2} allows an increase in J_c that depends sensitively on the applied field. At 20 T, I_c/I_{c0} lies between 3 and 4, compared to $I_c/I_{c0} \approx 2$ at 14 T. The change in ρ_n has an exponential dependence on fluence, so the rate of change of I_c/I_{c0} and B_{c2} with fluence eventually decreases. At higher fluences, the degradation of T_c owing to atomic disordering can reach 20% or more (§2), and this effect will eventually lower J_c . For alloyed wires, the initial enhancement of ρ_n with irradiation is smaller than for binary wires, owing to the higher initial value of ρ_n for the alloyed wires. In binary wires, an enhancement of $J_c(B)$ in measurements in high fields after irradiation is correlated to the introduction of radiation-induced flux pinning centers as ρ_n increases, but in alloyed wires, this effect is reduced or even suppressed (Ta addition). However, Weiss et al. did not advance an explanation for the decrease in J_c below initial values in alloyed wires at relatively low irradiation fluences of about 1 to $2 \times 10^{22}/\text{m}^2$ ($E = 14$ MeV).

Further comparative investigations of commercial binary and ternary Nb_3Sn conductors are discussed below, in §3.4 on high energy neutron irradiation. The general conclusions that ternary material is more radiation-sensitive were confirmed (Figures 3.4.7 and 3.4.8).

3.4. HIGH ENERGY NEUTRON IRRADIATION

Comparative neutron irradiations at 14 MeV of alloyed and binary Nb_3Sn were discussed above [Weiss et al., 1987]. Earlier results by Scanlon and Raymond [1979] on two types of bronze processed binary material are presented in Figure 3.4.1. The specimens were irradiated near 4 K with high energy neutrons from two sources with the spectra shown in Figure 3.4.2. These measurements were made at 4 K in fields of 12 T on (A) laboratory monofilament

Figure 2.1.5. T_c vs. neutron fluence. $\square$ —data from Bett [1974]. Δ —data from Schweitzer and Parkin [1974] and Sweedler et al. [1974]. Figure from Karkin et al. [1976].

Figure 2.1.6. Superconducting transition curves for Nb₃Sn diffusion wires before and after 4.6-K irradiation with fast neutrons and after annealing. From Soell et al. [1975].

2.1.2. Cryogenic-Temperature Irradiation

Apparently, the first measurements of T_c after cryogenic neutron irradiation were made by Soell et al. [1975] in the low temperature facility of the Garching reactor. Nb_3Sn diffusion wires were produced by vapor-phase diffusion of Sn into Nb wires of 0.3-mm diameter at 950°C for 24 h. A typical J_c at 5 T at 4.2 K for this material is 3000 A/mm² as compared to 6000 to 10 000 A/mm² for commercial material at that time. After irradiation at 4.6 K, specimens were moved from the irradiation position to the test cryostat without warmup. The results after irradiation to a fast neutron fluence of $3.9 \times 10^{22}/m^2$ ($E > 0.1$ MeV) are shown in Figure 2.1.6. Some later fluence measurements [Wallner et al., 1988] indicated that this fluence determination could be too low by a factor of ~1.3 [Coltman et al., 1981]. A T_c reduction of 0.8 ± 0.1 K was observed for this fluence. Annealing for 10 min at 250 K produced a further small decrease of T_c . After ambient-temperature irradiation to $10^{22}/m^2$ ($E > 1$ MeV) = $4 \times 10^{22}/m^2$ ($E > 0.1$ MeV), Sweedler et al. [1974] reported a similar decrease in T_c (Figure 2.1.3). This indicated that changes in superconducting properties of Nb_3Sn after ambient-temperature irradiation may be similar to effects of 4-K irradiation. In fact, this early indication was borne out by later J_c studies discussed below [e.g., Hahn et al., 1991], although Brown et al. [1978b] reported some significant differences in damage at different irradiation temperatures on certain specimen types (§3.2.3).

Söll et al. [1976] used similar diffusion specimens to extend their cryogenic irradiations to a fast neutron fluence of $\sim 10^{23}/m^2$ ($E > 0.1$ MeV). The actual irradiation temperature was ~10 K. A slight extrapolation of the results shown in Figure 2.1.7 indicates that a fluence of $\sim 1.2 \times 10^{23}/m^2$ would be required to reduce T_c to 13.5 K, the value obtained by Sweedler et al. [1974] after a higher ambient-temperature fluence of $5 \times 10^{22}/m^2$ ($E > 1$ MeV) or $2 \times 10^{23}/m^2$ ($E > 0.1$ MeV). This is a further indication of the very approximate equivalence of ambient and ~10-K irradiation in producing this type of damage in a superconducting property of Nb_3Sn . Further support for this equivalence is given in Figure 2.1.8, in which Söll et al. have plotted T_c vs. neutron or O-ion fluence in several experiments, and the data fall on three curves corresponding to the three different energies of the irradiating particles,

Figure 2.1.7. T_c vs. fast neutron fluence ($E > 0.1$ MeV) for an Nb₃Sn diffusion wire. From Söll et al. [1976].

Figure 2.1.8. T_c vs. O-ion or fast neutron fluence for several Nb₃Sn irradiation experiments. From Söll et al. [1976].

Figure 2.1.9. Decrease of T_c vs. mean energy transfer per lattice atom for the same irradiation experiments depicted in Figure 2.8. From Söll et al. [1976].

rather than to the temperature of irradiation. By calculating the mean energy transferred to each lattice atom by elastic collisions of the irradiating particle, and plotting the data as a function of this mean damage energy, E_D (E_p in figure), Söll et al. were able to bring the data from six different experiments into even greater correspondence, as shown in Figure 2.1.9.

No noticeable reduction in the T_c of the diffusion specimens was found up to a "threshold" fast neutron fluence of $10^{22}/\text{m}^2$ ($E > 0.1$ MeV), and then a linear decrease with fluence was observed by Söll et al. However, Ghosh et al. [1978] irradiated ~ 700 -nm (~ 7000 -Å) thick films of Nb_3Sn (prepared by co-evaporation of Nb and Sn) with 2.5-MeV α particles at 30 K and then measured T_c and ρ_n at lower temperatures. A plot of T_c vs. ρ_n (ρ_0) for three specimens shows that T_c does not decrease significantly until $\rho_n \approx 30 \mu\Omega\text{cm}$ (Figure 2.1.10). Ghosh et al. therefore inferred that the rate of depression of T_c with irradiation fluence was dependent on the initial state of the specimen. If the initial $\rho_n \approx 15 \mu\Omega\text{cm}$, T_c will drop relatively slowly. However, if the initial $\rho_n \approx 40 \mu\Omega\text{cm}$ ($T_c \approx 17$ K), Figure 2.1.10 (open circles) shows that T_c will drop rapidly with fluence. Viswanathan et al. [1978] also found that the initial degradation of T_c in V_3Si under radiation was sample-dependent. Ghosh et al. noted further that it was customary to use a logarithmic scale for the fluence, whereas, when the T_c data were plotted vs. a linear fluence scale, no "threshold" was observed and T_c essentially had a positive curvature with fluence. A theoretical analysis was advanced to explain these features of the data. Viswanathan et al. [1978] also reviewed theoretical analyses. From the standpoint of the practical requirements for the ITER TF magnets, the existence of a "threshold" is probably moot, in that very little change in T_c should be expected if the total fast neutron fluence remains below $10^{22}/\text{m}^2$. However, if the ρ_n of the chosen conductor is quite high, as is often the case for the newer ternary Nb_3Sn conductors, more sensitivity might be expected and the material should be tested after irradiation. Probably, screening tests could be done with ambient irradiation.

Brown et al. [1978a] also measured both ρ_n and T_c , but used fast neutron irradiation just above T_c (≈ 20 K). A decrease of about 1 K was observed after a fluence of $10^{22}/\text{m}^2$ ($E > 0.1$ MeV), as indicated in Figure 2.1.11. Since the initial ρ_n was $16 \mu\Omega\text{cm}$, and ρ_n at $10^{22}/\text{m}^2$ was $\sim 19.5 \mu\Omega\text{cm}$, these neutron-

Figure 2.1.10. T_c vs. ρ_n (ρ_0) for Nb_3Sn (and Nb_3Ge). From Ghosh et al. [1978].

irradiation results show a somewhat more rapid decline in T_c vs. ρ_n than the α -particle results of Ghosh et al. on films (Figure 2.1.10). The Nb_3Sn specimen used by Brown et al. to obtain the data in Figure 2.1.11 was also a film (thickness, 1620 nm).

2.2. COMPOSITE, BINARY Nb_3Sn

2.2.1. Ambient-Temperature Irradiation

Ostenson et al. [1983] used the HFBR to examine Nb_3Sn -Cu composites prepared by an in situ method, and compared the results to those on bulk specimens irradiated earlier in this reactor. In situ wire is prepared by casting an Nb-Cu billet, reducing the billet to wire, coating the wire with Sn, and annealing the wire at 550 to 650°C to form Nb_3Sn [Verhoeven et al., 1983]. The superconducting filaments in these specimens were small (~70-nm thick) and discontinuous. At 12 T, the critical current density, J_c , was 600 A/mm², which is comparable to recent strand grade specifications (Table 1.3.1). The temperature of irradiation was estimated to be ~80°C. Figure 2.2.1 shows that T_c remained approximately constant to a fluence of about $2 \times 10^{22}/m^2$ ($E > 1$ MeV), but decreased by almost 50% at $1.3 \times 10^{23}/m^2$, in approximate accord with the earlier results of Sweedler et al. on bulk material, also shown in the figure. The difference in response between two in situ processed samples is indicated by the open and filled symbols in Figure 2.2.1. The superconducting-to-normal transitions for these same samples were also measured by an ac susceptibility technique in which currents were induced around the circumference of the wire perpendicular to the long axis of the filaments. This required that the measuring current cross through the bronze barriers separating the filaments so this susceptibility technique measured the onset of appreciable proximity coupling. Measuring fields of 0.1 mT at 100 Hz were used. The samples did not show shielding against magnetic flux entry until temperatures approximately 4 K lower than the onset of superconductivity in the filaments were reached.

Figure 2.1.11. Changes in T_c and ρ_n (ρ) of Nb₃Sn vs. fast neutron fluence for 20-K irradiation. From Brown et al. [1978a].

Figure 2.2.1. T_c vs. fast neutron fluence ($E > 1$ MeV) measured by both susceptibility and resistivity techniques. Data from other investigations are also shown. From Ostenson et al. [1983].

Figure 2.2.2. T_c vs. fast neutron fluence for three Nb_3Sn materials and earlier results. From Kodaka et al. [1985].

Bronze processed, in situ, and powder metallurgy Nb₃Sn specimens prepared on a laboratory scale were reactor-irradiated at ~333 K together with commercial tapes. These specimens were subsequently stored at ambient temperature for long periods before measurements were made [Kodaka et al., 1985]. The results shown in Figure 2.2.2 include comparisons with the earlier ambient-temperature reactor results by Sweedler et al. [1974] and Ostenson et al. [1983]. The initial differences in degradation are small for the Nb₃Sn composites prepared by different methods, but at higher fluences, the in situ, tape, and bronze processed materials appear to fall significantly below the line established by Sweedler et al. for bulk Nb₃Sn. There is also significant disagreement between the results for the in situ material from two different sources.

Additional results of ambient- and above-ambient temperature irradiations with the RTNS 14-MeV source and a 30-MeV Be(d,n) source, as well as a fission reactor (HFBR), are shown in Figure 2.2.3 [Snead et al., 1981]. These results were obtained on both multifilamentary and single-core specimens fabricated by the bronze process. The data are plotted for equivalent HFBR fluences based upon damage-energy cross sections. Even when so plotted, the RTNS damage in the 19-core specimens at about $10^{23}/\text{m}^2$ (equivalent) is higher than expected, suggesting that the damage-energy scaling underestimates the disorder produced in this type of specimen, though no anomaly is seen in the single-core specimen results. The high energy ($E > 5$ MeV) fluence at the ITER TF magnets may be about $1.5 \times 10^{21}/\text{m}^2$ [Sawan, 1993] for a total fast-neutron fluence of $10^{22}/\text{m}^2$ ($E > 0.1$ MeV). The fluence in Figure 2.2.3 equivalent to this high energy fluence of $1.5 \times 10^{21}/\text{m}^2$ would still be well below the region ($>10^{23}/\text{m}^2$) where significant damage from the RTNS fluence was observed. However, comparative data on lower-fluence RTNS irradiation and fission-reactor irradiation of multifilament specimens are lacking.

2.2.2. Cryogenic-Temperature Irradiation

Scanlon and Raymond [1979] used both the RTNS source of 14-MeV neutrons and a Be(d,n) source with a significant component above 4 MeV (extending to 30 MeV) to irradiate both monofilamentary and multifilamentary wires fabricated by the bronze process. The irradiations were performed at 4.2 K. No change in T_c within the measurement accuracy of ± 0.5 K was observed, at the highest

Figure 2.2.3. Normalized T_c of single-core and 19-core wires vs. corrected fast neutron fluence ($E > 0.11$ MeV). RTNS irradiation results for Nb_3Sn are plotted vs. the equivalent HFBR fluence. Horizontal error bars represent fluence uncertainties. From Snead et al. [1981].

fluence of $\sim 7 \times 10^{21}/\text{m}^2$. Since this 14-MeV fluence may be equivalent to a fast neutron fluence of about $4 \times 10^{22}/\text{m}^2$ ($E > 0.11$ MeV) [see Figure 3.4.6(B) below], the lack of correspondence with the results of Brown et al. [1978a] on bulk film specimens is somewhat surprising, but perhaps explicable by the inaccuracy of the measurements and the fact that the fluence is near the threshold for observable T_c change.

Guinan et al. [1984] irradiated similar bronze processed, monofilamentary Nb_3Sn wire at 4.2 K with 14-MeV neutrons from the RTNS source. Some temperature excursions to a maximum of 61.5 K occurred during the cryogenic irradiation, but were not expected to result in significant annealing. These results were compared to the previous data on these specimens from both fission and high-energy neutron sources with E_p , the calculated damage energy. Figure 2.2.4 shows that ΔT_c can be correlated fairly well by this method. See also Snead et al. [1976] for an explanation of this correlation method. Snead et al. found that T_c changes correlated well with E_p although J_c changes did not.

2.3. COMPOSITE, TERNARY Nb_3Sn

2.3.1. Ambient-Temperature Irradiation

Measurements on several types of both binary and alloyed Nb_3Sn multifilamentary wires were made after ambient-temperature irradiations in the 14-MeV RTNS source [Weiss et al., 1987]. Similar, linear decreases of T_c with fluence were observed for all specimens (Figure 2.3.1). The highest 14-MeV fluence of $\sim 3.3 \times 10^{22}/\text{m}^2$ is roughly equivalent to a reactor fluence of $2 \times 10^{23}/\text{m}^2$ ($E > 0.11$ MeV) [see Figure 3.4.6(B) below], so the substantial decreases in T_c observed at the highest fluence are in agreement with previous work with lower-energy fluences. Specimen A in Figure 2.3.1 had 10 000 cores and specimen B had 19. The depression in T_c at a fluence of $3.3 \times 10^{22}/\text{m}^2$ was larger by about 0.5 K for the alloyed wires, indicating some difference in the damage mechanism. In binary Nb_3Sn , irradiation is thought to result in mostly homogeneous antisite disordering by replacement collisions, as explained further in §§3 and 5 below. In ternary, or alloyed wires, the alloying of Ta has been shown to enhance the sensitivity of T_c to strain by a factor of two

Figure 2.2.4. ΔT_c vs. damage energy, E_D , for bronze-processed monofilamentary Nb_3Sn for irradiations at: Δ -HFBR (60°C), $\square$ -RTNS-I (20°C), $\circ$ -30 MeV d-Be, and $\blacksquare$ -RTNS-II (4.2 K). From Guinan et al. [1984].

Figure 2.3.1. T_c for different binary and alloyed Nb_3Sn multifilamentary wires vs. 14-MeV neutron fluence at 25°C. From Weiss et al. [1987].

Table 2.3.1. Commercial Nb₃Sn Sample Specifications.
From Hahn et al. [1991].

Parameter	SEWC	SUMI	TWC-247	TWC-246	IGC	TWC-325
Conductor diam. (mm)	0.193	0.200	0.193	0.193	0.330	0.193
Number of filaments	745	1	850	815	330	820
Filament diam. (μm) –	3.4	91	2×4	2×4	4.0	2×4
Cu-stabilizer (vol%)	31.6	32.0	33.9	33.9	53.0	26.0
Diffusion barrier	Nb	Nb	V	V	Ta	V
Filamt. alloy (wt%)	Nb	Nb	Nb	Nb–0.8 Ti	Nb–1.1 Ti	Nb–2.0 Ti
Sn core alloy (wt%)	Sn	Sn	Sn–3.2 Ag	Sn–3.2 Ag	Sn	Sn–0.7 Cu
Nb ₃ Sn (vol%)	20.0	20.7	23.2	22.3	16.3	22.4

[citation, Weiss et al., 1987]. By analogy, Weiss et al. suggested that an additional process capable of influencing compressive strains should be invoked to explain the faster decrease of T_c with fluence for ternary Nb₃Sn superconductors.

2.3.2. Cryogenic-Temperature Irradiation

An additional RTNS experiment at both 14 K and ambient temperature on pure and alloyed multifilamentary Nb₃Sn conductors was carried out on the composite conductors characterized in Table 2.3.1 [Hahn et al., 1991]. Figure 2.3.2 shows the decrease in T_c for binary specimens and Figure 2.3.3 shows the decrease for Ti-alloyed conductors as function both of damage energy and fluence. Equations describing the depression in T_c are given in the figures. Depressions (extrapolated slightly) of about 3 K or less for binary Nb₃Sn at 14-MeV fluences of $\sim 2.5 \times 10^{22}/\text{m}^2$ are in approximate agreement with the results of Weiss et al. [1987], as are the slightly larger decreases of up to 4 K for ternary Nb₃Sn. However, Figure 2.3.1 from Weiss et al. shows somewhat smaller decreases in T_c at higher 14-MeV fluences than those employed by Hahn et al. No pronounced differences were observed between ambient-temperature and low temperature results. The results were also expressed as

$$[T_{\text{com}}(E_D)/T_{\text{com}}(0)] = \exp(-E_D/4), \quad (2.3.1)$$

where E_D is in eV/atom (for 14-MeV neutrons, 1 eV/atom = $1 \times 10^{22}/\text{m}^2$) and the results apply to the unstrained wires [Summers et al., 1991].

The initial rate of decrease of T_c with irradiation dose was higher for materials with lower initial T_c ; the range of decrease varied from 4.4 K/eV (SUMI) to 6.5 K/eV (TWC-325). As the Ti concentration of the ternary conductors increased, the initial T_c decreased and the rate of decrease in T_c with irradiation also increased. Hahn et al. suggested that both of these Ti-effects arose from initial strain. They noted that the lattice parameter had been found to increase by 0.1%/K of T_c depression in both foils and multifilamentary specimens [citations, Hahn et al., 1991].

Since the maximum 14-MeV fluence (or, slightly extrapolated fluence—see curves) in these experiments was $\sim 2.5 \times 10^{22}/\text{m}^2$, equivalent to a fast neutron fluence of $\sim 1.4 \times 10^{23}/\text{m}^2$ ($E > 0.1$ MeV) [see Figure 3.4.6(B) below], these

Figure 2.3.2. T_c vs. 14-MeV neutron fluence or damage energy for in situ (full symbols) or ambient-temperature (closed symbols) irradiated conductors fitted with negative exponential equations. Characteristics of the commercial Nb_3Sn conductors are given in Table 2.3.1. All conductors in this graph are binary. From Hahn et al. [1991].

Figure 2.3.3. T_c vs. 14-MeV neutron fluence or damage energy for in situ (full symbols) or ambient-temperature (closed symbols) irradiated conductors fitted with negative exponential equations. Characteristics of the commercial Nb_3Sn conductors are given in Table 2.3.1. All conductors in this graph are ternary: 0.8 wt.% Ti (TWC-246), 1.2 wt.% Ti (IGC), and 2.0 wt.% Ti (TWC-325). From Hahn et al. [1991].

Figure 2.4.1. T_c vs. annealing temperature for 1-h anneals of irradiated Nb_3Sn in pure He. From Voronova et al. [1978].

results appear to be consistent with the earlier literature. For example, Bauer et al. [1975] found about a 7-K decrease of T_c at $5 \times 10^{22}/\text{m}^2$, but this fluence was quoted for $E > 0.82$ MeV. Ostenson et al. [1983] found a decrease in T_c of about 7 to 8 K at $\sim 1 \times 10^{23}/\text{m}^2$, but since $E > 1$ MeV for this fluence, the equivalent fluence with $E > 0.1$ MeV could be as high as $4 \times 10^{23}/\text{m}^2$.

2.4. RECOVERY OF THE TRANSITION TEMPERATURE

2.4.1. Bulk Specimens

Some early data presented in Figure 2.1.1 showed that an annealing temperature between 750 and 900°C was required to restore T_c . This result has been corroborated by other investigations. Figure 2.4.1 from Voronova et al. [1978] shows the results of 1-h anneals at the indicated temperatures on diffusion specimens irradiated with α particles and protons at 100°C. Figure 2.4.2 from Karkin et al. [1976] shows a similar behavior for the recovery of T_c after ambient-temperature, fast neutron irradiation of vacuum-annealed Nb and Sn pressed powder specimens. Recovery is complete at 900 to 950°C, optimum temperatures for Nb₃Sn ordering. The isochronal annealing was carried out in pure He for periods of 20 min. Both resistance and inductance measurements of T_c coincided.

2.4.2. Composite Specimens

Figure 2.4.3 from Kodaka et al. [1985] shows the recovery process for in situ Nb₃Sn wires and multifilamentary Nb₃Sn wires produced by the bronze process. These specimens were also neutron-irradiated ($E > 0.1$ MeV) near ambient temperature at ~ 333 K. For these more modern materials, complete recovery seemed to require somewhat higher annealing temperatures.

2.4.3. Cryogenic Recovery

Brown et al. [1977b, 1978a] irradiated an Nb₃Sn film to a fast neutron fluence of $1.32 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV) at 20 K, and measured a decrease in T_c to 16.7 K. Isochronal annealing showed a small recovery stage beginning at about 100 K and a larger recovery stage beginning at about 270 K and extending up to

Figure 2.4.2. T_c vs. isochronal annealing temperature for Nb_3Sn specimens irradiated to fluences of $F \times 10^{23}/m^2$. $\times$ - $F = 2$; Δ - $F = 4$; O - $F = 7$. From Karkin et al. [1976].

A

B

Figure 2.4.3. T_c vs. annealing temperature for in situ (A) and bronze process (B) Nb₃Sn material irradiated to neutron fluences of 4.25 (A) and 3.14 (B) $\times 10^{23}/\text{m}^2$. From Kodaka et al. [1985].

Figure 2.4.4. T_c and $\rho_n(\Delta\rho)$ during annealing of neutron-irradiated Nb_3Sn film. From Brown et al. [1978a].

500 K (Figure 2.4.4). The lower-temperature recovery was attributed to interstitial (Frenkel-pair) migration and the broader, higher temperature recovery was attributed to elimination of antisite disorder [Snead and Luhman, 1986]. See also §5.1.

A model for the recovery process developed by Dew-Hughes et al. [1978] was based on recovery data from a number of A-15 compounds. At low temperatures, the recovery was assumed to take place by vacancy assisted reordering, but at high temperatures, recovery was ascribed to the motion of thermal equilibrium vacancies. Thus, a theoretical foundation for the recovery process exists. The nature of the A15 defects that must recover is discussed further in the section on the order parameter, §5. For design purposes, however, the corroboration of the recovery temperature range by several different groups is more important than theoretical speculation.

2.5. EFFECTS OF STRAIN

Snead and Suenaga [1980] investigated the synergism of irradiation damage and strain on single-core wires of Nb in Cu-13% Sn reacted at 700°C for 4 d. The bronze-to-Nb ratio was 15:1 and the Nb₃Sn layer thickness was 6 μ m for a wire diameter of 0.35 mm. The T_c was measured resistively. Neutron irradiation in the HFBR at ~100°C showed that the change in T_c owing to the prestrain was enhanced for fluences above about $10^{22}/\text{m}^2$ ($E > 1.0$ MeV). The maximum enhancement was 0.8 K or 200% of the total decrease in T_c for an unirradiated wire. The results of the measurements with and without the bronze are shown in Figure 2.5.1. The two points on the Y axis show the difference before irradiation; this difference increased with fluence and was attributed to the radiation enhancement of the "bronze effect" of prestrain. There is evidence that radiation damage in A-15 compounds results in off-line "static displacements" of atoms on the A chains, perhaps from outward buckling of the A chains adjacent to a disordered site with a vacancy or a B atom on an A site. The buckling increases the atomic separation, decreasing the atomic overlap and local density of states, and, hence, T_c . The application of a compressive stress results in an additional outward buckling and further T_c reduction. The synergism of radiation damage and prestrain is negative unless

Figure 2.5.1. Comparison of T_c vs. fast neutron fluence ($E > 1$ MeV) for single-core Nb_3Sn wire with and without bronze. Irradiation temperature was $\sim 100^\circ\text{C}$. From Snead and Suenaga [1980].

externally applied tensile strain on the conductor counterbalances the prestrain.

Summers et al. [1991] expressed the zero-field critical temperature, T_{c0} , as a function of strain and damage energy from irradiation:

$$T_{c0}(\epsilon, E_D) = T_{c0m} \exp(-E_D/4) [1 - a |\epsilon_M + \epsilon(E_D)|^u]^{1/w}, \quad (2.5.1)$$

where $T_{c0m} = 18.3$ K, $u = 1.7$, $w = 3$, ϵ_M is the mechanical strain, and $a \approx 900$ for $\epsilon < 0$, $a \approx 1250$ for $\epsilon > 0$. The parameter ϵ refers to the intrinsic strain, ϵ_0 , as defined in §1.4. A comparison of this equation with experimental data had not been published at this writing.

Figure 3.1.1. I_c vs. transverse field before and after neutron irradiation for specimens (A) with low initial I_c and (B) with higher initial I_c . From Cullen and Novak [1966].

3. RADIATION-INDUCED CHANGES IN THE CRITICAL CURRENT

In comparing radiation results on J_c from different sources, it is important to note that J_c is strongly dependent upon the magnetic field in which it is measured. The first figure in this section illustrates this dependence.

3.1. BULK, BINARY Nb_3Sn

3.1.1. Ambient-Temperature Irradiation

Studies of radiation effects on the J_c of Nb_3Sn were started about 30 years ago. The early results of Cullen and Novak [1966] on vapor-deposited strip material (2×10^{-5} cm² in cross section) foreshadowed later results, for they found that irradiation to a fast neutron fluence of $1.4 \times 10^{22}/m^2$ increased the J_c of material that had a low initial J_c . For material of higher initial J_c , a sharp decrease in J_c was observed as the irradiation fluence was increased from 3.4 to $7.2 \times 10^{21}/m^2$, and even a 1-h anneal at 800°C only partially restored the original J_c . These features are shown in parts A and B of Figure 3.1.1, which depict I_c vs. transverse applied field after fast neutron irradiation at -50°C and a three-to-four week storage period for decay of radioactivity. Proton irradiation (mean energy 2.83 MeV) and deuteron irradiation were also shown to initially increase the J_c of Nb_3Sn diffusion layers on an Nb tube [Bode and Wohlleben, 1967; Wohlleben, 1969].

Figure 3.1.2 presents the results of proton irradiation from 100 to 200°C on J_c as measured in fields up to 5 T. In general, the increase in J_c was somewhat stronger for lower initial J_c . The initial increase in J_c appeared to be independent of the applied field. This increase was explained by the formation of new flux pinning centers formed by the atoms displaced during irradiation. Material with an initially high J_c already has a high density of pinning centers (§1.1), and therefore, is less susceptible to improvement with irradiation. Complete recovery required a temperature of ~800°C, a temperature region in which reordering is expected.

Irradiation by 24-MeV O ions at ambient temperature was also shown to increase J_c , except at higher fluences [Ischenko et al., 1972]. These results on diffusion layers of Nb_3Sn are shown in Figure 3.1.3.

Figure 3.1.2. Enhancement of J_c per cm tube length for Nb₃Sn diffusion layers vs. 2.83-Mev proton fluence. (a) shows measured values at 5 T; (b) shows mean values at 1.2, 3, and 5 T. From Bode and Wohleben [1967].

Figure 3.1.3. J_c vs. transverse field for unirradiated and O-ion irradiated Nb_3Sn diffusion-layer specimens. From Ischenko et al. [1972].

Figure 3.1.4. I_c vs. transverse field for Nb₃Sn diffusion wires before and after fast neutron irradiations at ambient temperature. From Bauer et al. [1975].

Bauer et al. [1975] also reported increases in J_c with fast neutron fluences up to $10^{22}/\text{m}^2$ ($E > 0.82$ MeV), but then a substantial decrease was observed at $5 \times 10^{22}/\text{m}^2$, except at higher magnetic fields (Figure 3.1.4). The specimens were diffusion wires and the irradiation temperature was thought to be not much above 80°C .

3.1.2. Cryogenic-Temperature Irradiation

The first low temperature irradiation studies of J_c were made with 15-MeV deuterons by Coffey et al. [1967] at 30 K on vapor-deposited Nb_3Sn . About 0.3% atomic displacements could be produced with the available fluence. In correspondence to the ambient-temperature irradiation results, large increases in the J_c of low J_c material and a decrease in the J_c of high J_c material were observed. These very different material-dependent responses are presented in Figure 3.1.5, parts A (initial, 0-field $J_c \approx 4000$ A/mm²) and B (initial, 0-field $J_c \approx 13\,000$ A/mm²). The large decrease in J_c in part B of Figure 3.1.4 corresponded to a drop in T_c of only 1 K, which equates very approximately to a fast neutron fluence of about $4 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV) (§2.1.2, [Soell et al., 1975]). Less than 25% of the induced effects were recovered by annealing to 300 K. Enhancement of the low J_c material increased with field strength, up to about 5 T.

Soell et al. [1975] irradiated Nb_3Sn diffusion wire at 4.6 K to a fast neutron fluence of $3.9 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV), but still did not observe a decrease in J_c , as Figure 3.1.6 indicates. Measurement fields were limited to 5 T. Owing to different irradiation species or spectra and unreported values, it is difficult to compare this work with earlier bulk specimen studies.

3.2. COMPOSITE, BINARY Nb_3Sn

3.2.1. Ambient-Temperature Irradiation

Cullen and Novak [1966] reported a decrease in J_c on bulk, high J_c material at a fast neutron fluence of $7.2 \times 10^{21}/\text{m}^2$ (E not specified), as indicated in Figure 3.1.1(B). Parkin and Schweitzer [1973] irradiated multifilamentary specimens produced by drawing a Cu-10% Sn billet containing 19 Nb rods to a

Figure 3.1.5. J_c vs. transverse field at (A) 5.7 K and (B) 10.9 K. — pre-irradiation, - - - 1.0×10^{21} deuterons/m² ($E = 15$ MeV), - · - · - post 77 K, and · · · · post 300 K. Nb₃Sn vapor-deposited specimens. From Coffey et al. [1967].

Figure 3.1.6. J_c vs. transverse field for Nb₃Sn diffusion wires before and after fast neutron irradiation and subsequent annealing. From Soell et al. [1975].

Figure 3.2.1. Normalized I_c at 4 T vs. transverse field before and after irradiation to a fast neutron fluence of $1.1 \times 10^{23}/\text{m}^2$ ($E > 1.0 \text{ MeV}$). Nb_3Sn 19-filament composite specimens. From Parkin and Schweitzer [1973].

dimeter of 0.4 mm. This composite was reacted at 700°C for 28 h to form a layer of Nb₃Sn on the Nb that was ~1.3 μm thick. A large decrease in J_c was seen after a fast neutron fluence of $1.1 \times 10^{23}/\text{m}^2$ (E > 1.0 MeV) from the BNL HFBR at 60 ± 5°C. The maximum field was 4 T (Figure 3.2.1).

Parkin and Sweedler [1975] irradiated similar multifilamentary composites in the HFBR at 60 ± 5°C over a wider fluence range. In contrast to some bulk Nb₃Sn studies, no enhancement of J_c was observed in the measurements in a 4-T field, even at lower fluences of $10^{21}/\text{m}^2$ (Figure 3.2.2). Rather, above a threshold fluence between 2 and $3 \times 10^{22}/\text{m}^2$ (E > 1 MeV), a very rapid reduction of J_c was observed. This threshold effect is also illustrated in Figure 3.2.3, where the critical current, I_c, is plotted vs. the magnetic field to 2 T. In subsequent ambient-temperature irradiation of specimens of the same material type by Snead and Parkin [1976] in the HFBR, the field strength in which the measurements were done was extended to 16 T (Figure 3.2.4). Again, decreases in J_c were observed at a fluence of $4.5 \times 10^{22}/\text{m}^2$ (E > 1 MeV). However, in agreement with some bulk specimen studies, small increases in J_c were observed at lower fluences. These increases were more pronounced at the higher field strengths. See also Figure 3.3.2 below, where some of these data are replotted for E > 0.1 MeV. The J_c reduction in the high fluence regime of this figure appears to be independent of field. This J_c reduction at high fluences [$>2 \times 10^{22}/\text{m}^2$ (E > 1 MeV) or $>8 \times 10^{22}/\text{m}^2$ (E > 0.1 MeV)] is usually ascribed to the decrease in T_c to ~3 K as J_c is measured at 4 K [Snead and Luhman, 1986].

In some of the earlier studies on bulk Nb₃Sn, an initial irradiation increase in J_c was more evident in non-optimized material that had a lower initial J_c (e.g., Figures 3.1.1 and 3.1.5). However, when Colucci et al. [1977] investigated both the earlier 19-filament composite wire used in many studies (type A) and an improved material with a 60% higher initial J_c (type B), the opposite result was found (see also Colucci and Weinstock [1978]). The type B material contained one Nb₃Sn filament of $5.7 \times 10^{-6} \text{ cm}^2$ cross sectional area, whereas the combined cross section of the multifilamentary material was $7.4 \times 10^{-6} \text{ cm}^2$. Both specimen types were prepared by using the surface reaction of Nb wire co-drawn in a Cu-Sn matrix at 700°C for 24 h. Higher values of the matrix Sn concentration and the volume ratio of the matrix to Nb were achieved

Figure 3.2.2. I_c at 4 T at 4.2 K vs. fast neutron fluence for Nb_3Sn multifilamentary composites. Irradiation at $\sim 60^\circ\text{C}$. From Parkin and Sweedler [1975].

Figure 3.2.3. I_c at 4.2 K vs. transverse field for Nb_3Sn multifilamentary composites irradiated with fast neutrons at $\sim 60^\circ\text{C}$. From Parkin and Sweedler [1975].

Figure 3.2.4. I_c of multifilamentary Nb₃Sn composites irradiated with fast neutrons at -60°C vs. transverse field. From Snead and Parkin [1976].

in the type B material. The irradiation response of material of types A and B is compared in Figure 3.2.5. The radiation enhancement of J_c in type B was fractionally larger than in type A. The irradiation degradation of type B in low fields was attributed to a reduction in T_c by atomic disordering which would be offset at larger fields by increased pinning forces.

Irradiation tests were also performed on Nb_3Sn composite conductors fabricated by the in situ rather than the bronze process [Ostenson et al., 1983]. In situ material is prepared by a casting and drawing technique (§2.2.1). The chief difference between the in situ and conventional bronze process material is that the superconducting filaments are much smaller (~70-nm thick), and discontinuous. Since this structure might alter the radiation susceptibility, irradiation at ~80°C in the HFBR was carried out. Both resistive and susceptibility measurements of T_c were made (§2). It was found that irradiation results were similar to those of bulk Nb_3Sn . For fluences up to $6 \times 10^{21}/m^2$, J_c and B_{c2} increased, presumably because of increases in ρ_n [Equation (3.2.2)]. At higher fluences, the damage was sufficient to lower T_c appreciably, and both J_c and B_{c2} were reduced. The results were found to be similar to results for single-core Nb_3Sn filaments reported earlier.

3.2.2. Cryogenic-Temperature Irradiation

Brown et al. [1975] irradiated multifilamentary, Cu-clad wires of Nb_3Sn at 6 K in the CP-5 reactor facility at ANL. In contrast to the ambient-temperature results of Bauer et al. [1975] and of Parkin and Sweedler [1975] which indicated that fluences of about $5 \times 10^{22}/m^2$ ($E > \sim 1$ MeV) would be required for substantial J_c decreases, the peak increases in J_c were observed at a fluence of $\sim 0.8 \times 10^{22}/m^2$ ($E > 0.1$ MeV) and decreases in J_c were observed after that point (Figure 3.2.6). At a fluence of $1.8 \times 10^{22}/m^2$, J_c had still not dropped below the initial value, except for measurements made in lower fields of 1.27 and 0.63 T. However, in a later publication Brown et al. [1978b] noted that these fluences should be multiplied by 2.2. This brings the data into better agreement with the earlier results at ambient temperature. Figure 3.2.7 shows J_c vs. magnetic field. Comparing this figure with the data of Soell et al. [1975] on bulk Nb_3Sn diffusion specimens (Figure 3.1.6) irradiated to a similar fast neutron fluence indicates a lack of agreement in

Figure 3.2.5. J_c vs. transverse field for two types of Nb_3Sn conductors after fast neutron irradiation ($E > 0.1 \text{ MeV}$) at $\sim 6 \text{ K}$. From Colucci et al. [1977].

Figure 3.2.6. Fractional change in J_c vs. neutron fluence and annealing temperature for fields from 0.63 to 3.32 T. Fluence given should be multiplied by 2.2. Nb_3Sn 19-filament composite specimens irradiated below 6 K. From Brown et al. [1975].

Figure 3.2.7. J_c vs. transverse field before and after fast neutron irradiation ($E > 0.1$ MeV) and after a 5-min anneal at 295 K. Fluence given should be multiplied by 2.2. Nb_3Sn 19-filament specimens irradiated below 6 K. From Brown et al. [1975].

the irradiation improvement of J_c . However, the much lower initial value of J_c in the material that Soell et al. used may have permitted a greater degree of irradiation improvement in J_c . About 25% of the change in J_c observed by Brown et al. recovered after an anneal at 295 K, in agreement with results of Coffey et al. [1967] discussed above (§3.1.2). This result also appears to contrast with data of Soell et al., who recorded a larger recovery after cryogenic irradiation of their bulk diffusion specimen to a similar fluence.

Colucci et al. [1976] carried out additional fast neutron irradiations in the CP-5 reactor at ~6 K on the same type of bronze processed Nb_3Sn composite wires (prepared by M. Suenaga of BNL) of 19 filaments co-drawn in a Cu-Sn matrix to a diameter of 0.25 mm and reacted at 700°C for 24 H. Although the irradiations were performed at ~6 K, specimens were stored in liquid N_2 as part of the procedure for transfer to the cryostat in which measurements were made. Annealing to 77 K was expected to change J_c by less than 20% [citations, Colucci et al., 1976]. Measurements were made in higher fields than the 3.3 T available to Brown et al. [1975]. At the higher fields up to 10 T (Figure 3.2.8), increases were observed in J_c up to the highest fluences of $1.1 \times 10^{22}/m^2$. However, a crossover in the curves was observed for low fields, so that the opposite effect of a decrease in J_c with irradiation is seen. The increase in J_c was attributed to the introduction of radiation-induced flux pinning centers. Colucci et al. suggested that an increase in J_c with fluence occurred until the potential wells associated with pinning centers began to overlap, and then subsequent irradiation decreased J_c . Also, J_c values for Nb_3Sn were considered not to be optimized, since most flux pinning occurs at grain boundaries, and the combined temperature dependence of the Nb_3Sn growth rate and the grain size result in a relatively large grain size. The radiation-induced shift in J_c is negative at low fields and positive at higher fields. The degradation of J_c results from a reduction in T_c owing to atomic disordering. However, this degradation is offset at larger fields by the increased pinning forces. (The pinning force is $J \times B$.) The additional depression in J_c below 4 T by annealing was considered somewhat surprising by Colucci et al., but was consistent with the results shown above in Figure 2.1.6 in which T_c was further decreased by a post-irradiation anneal.

Figure 3.2.8. J_c vs. transverse field for several irradiation fluences and annealing temperatures. Nb₃Sn 19-filament composite specimens. From Colucci et al. [1976].

Takamura and Kato [1981] also irradiated single-core and multifilamentary Nb₃Sn specimens at cryogenic temperatures (~5 K). They observed no enhancement in J_c in their single-core specimens to 6 T and a small enhancement in their 19-filament specimens after a fast neutron fluence of $3 \times 10^{21}/\text{m}^2$ (E > 0.1 MeV).

3.2.3. Comparison of Irradiation Temperature and Material Variations

A comparison of the cryogenic irradiation results of Brown et al. [1975] with the ambient irradiation results of Parkin and Sweedler [1975] on the same material in fields to about 4 T shows that no J_c increase was observed after ambient irradiation. (Compare Figure 3.2.2 with Figure 3.2.6.) This disagreement of ambient-temperature results with the low temperature irradiations of this type of material were addressed by Brown et al. [1977a], who extended the dose in 8-K irradiations by using thermal-neutron irradiation of ²³⁵U-doped specimens. One thermal neutron was considered equal to 20 ± 3 fast neutrons, owing to additional fission-fragment damage from the ²³⁵U. The equivalence was derived from the T_c change reported by Söll et al. [1976] after cryogenic fast neutron irradiation. (E was not specified by Brown et al.) Figure 3.2.9 shows the same behavior observed earlier by this group for fast neutron irradiations at lower fluences (see Figure 3.2.6). Thus, the higher fluence used by Parkin and Sweedler was considered not to explain the earlier dissimilarity in results with those of Brown et al. [1975]; rather, this dissimilarity was attributed to the difference in irradiation temperature. Brown et al. suggested that the dissimilarity was due to the decreased flux pinning by the microstructure (dislocation loops) that remained after the irradiation. Ambient irradiation near 400 K where some of the defects are mobile results in defect annihilation and agglomeration so that the final defect structure was capable of less flux pinning than the defect cascade structure that results from the same neutron fluence at low temperature.

Brown et al. [1978b] performed additional fast neutron irradiations to study further the effects of irradiation temperature and initial J_c described above. Two sets of monofilamentary samples were prepared by the bronze process at 700°C for 20 h (A) or at 800°C for 6 h (B). The longer heat treatment (A)

Figure 3.2.9. Normalized J_c vs. equivalent fast neutron fluence in several transverse fields for multifilamentary Nb_3Sn composites after irradiation at 6 K. From Brown et al. [1977a].

resulted in a higher J_c owing to the smaller grain sizes. Specimens were irradiated at both 6 and 350 K ($E > 0.1$ MeV). (Recalibration of the flux indicated that the fluences given by Brown et al. [1975] should be multiplied by 2.2 for comparison with this work.) Figures 3.2.10 and 3.2.11 show that there is a strong effect of irradiation temperature on the type A high J_c material (2×10^4 A/mm², 5 T), but virtually no effect on the low J_c material (8×10^3 A/mm², 5 T). The magnitude of the change in J_c with irradiation is more pronounced for the low J_c material, in agreement with earlier results, except for those of Colucci et al. [1977]. As explained previously, this difference is thought to result from irradiation-induced defects that enhance flux-pinning capability. In the high J_c material (A), the damage cascades resulting from the low temperature irradiation were thought to be much better flux pinners than the clusters from the high temperature irradiation. Thus, the effects of irradiation temperature and fluence on J_c were shown to be strongly dependent upon characteristics of the specimens.

Maier and Seibt [1978] also demonstrated that the material type affected radiation response in Nb₃Sn. They irradiated single core Nb₃Sn bronze processed specimens prepared at reaction temperatures between 650 to 850°C. Figure 3.2.12 shows the variability in the results after irradiation with 50-MeV deuterons at 18 K. These measurements were made at 4.2 K in a transverse field of 7 T.

Takamura and Okuda [1978] also demonstrated a material-based response when they irradiated Zr- and Ge-doped Nb₃Sn diffusion wire with fast neutrons at about 5 K in the LHTL (Liquid Helium Temperature Loop) at JAERI (Japan Atomic Energy Research Institute). The measurements, made at 4.2 K without warm up, are shown in Figure 3.2.13. The critical current increased in the Zr-doped samples but decreased in the Ge-doped samples after a fluence of $3.2 \times 10^{21}/\text{m}^2$ ($E > 0.1$ MeV).

Despite these variations observed for different Nb₃Sn samples, an overall uniformity of response was reported by Söll [1978] in a review. Söll compared the results of irradiations with different particle types by using the parameter E_D which represents the mean energy transferred by a given irradiation fluence to a lattice atom by an elastic collision that produces a

Figure 3.2.10. Normalized J_c vs. fast neutron fluence for high J_{c0} material after (A) 6-K or (B) 350-K irradiation and after 5-min anneals. Nb_3Sn multifilamentary specimens. From Brown et al. [1978b].

Figure 3.2.11. Normalized J_c vs. fast neutron fluence for low J_{c0} material after 6-K or 350-K irradiation and after 5-min anneals. Nb_3Sn multi-filamentary specimens. From Brown et al. [1978b].

Figure 3.2.12. Normalized J_c in a 7-T transverse field vs. 50-MeV deuteron fluence for single-core Nb_3Sn . Irradiation at 18 K. From Maier and Seibt [1978].

Figure 3.2.13. I_c of Nb₃Sn diffusion wire doped with (1) Ge or (2) Zr after ~5-K fast neutron irradiation to $3.2 \times 10^{21}/\text{m}^2$. From Takamura and Okuda [1978].

Figure 3.2.14. J_c normalized to the maximum value produced by irradiation vs. mean energy E_D (E_p) transferred to an atom for different Nb_3Sn irradiation experiments. Most irradiation was at ambient temperatures. See Söll [1978] for citations. The equivalent fast neutron fluence is for $E > 0.1$ MeV. From Söll [1978].

displacement. The equivalent fast neutron fluence ($E > 0.1$ MeV) is also shown in Figure 3.2.14, which gives results from eight different investigations (publication dates, 1969 to 1977). The specimens included both bulk and composite material. Irradiations were carried out mostly at ambient temperatures, but two cryogenic irradiation studies were included. The normalized J_c values that were compared in this figure were measured at a field of 5 T (or, at the highest applied field used in the experiment).

Snead [1977] has disputed the flux pinning mechanism usually invoked as the explanation for the increase in J_c . When the irradiations were carried out at low temperature, a substantial amount (~25%) of the observed increases in J_c were recovered upon annealing to ~300 K [Brown et al., 1976; Colucci et al., 1976]. However, Snead proposed that defects large enough to be flux pinners should not recover at room temperature. Rather, only Frenkel pairs and small defects should recover. The critical current, I_c , can be expressed as

$$I_c(B) = B_{c2}^n f(B/B_{c2}), \quad (3.2.1)$$

where f and the exponent, n , depend upon the size and nature of the flux-pinning sites. Also, B_{c2} can be expressed in the "dirty limit" as

$$B_{c2} = 3.06 \times 10^4 \gamma T_c \rho_n, \quad (3.2.2)$$

where γ (erg·cm⁻³·K⁻²) is the electronic specific heat coefficient and B_{c2} is in units of gauss. Since γ and T_c do not change rapidly at lower fluences, the increases in B_{c2} (where I_c approaches zero) seen in the figure of Snead and Parkin [1976] (Figure 3.2.4) were attributed to increases in ρ_n . The increase in B_{c2} in Equation (3.2.1) then accounts for the increase in I_c values in the low fluence regime. The contribution to ρ_n from Frenkel pairs and small defect clusters would make I_c changes at low temperature larger than those for ambient-temperature irradiations. Snead then attributed the increases in I_c at low fluences to increased B_{c2} owing to ρ_n increases, with flux-pinning mechanisms from large radiation-induced defects such as dislocation loops playing an undetermined, but much smaller, role.

3.3. TERNARY (ALLOYED), Nb₃Sn

Several investigations of the radiation response of alloyed Nb₃Sn samples have demonstrated a clear difference in sensitivity compared to that of binary, or

Figure 3.3.1. Normalized J_c vs. damage energy (E_D) for an alloyed Nb_3Sn conductor irradiated at 5 K compared to binary Nb_3Sn irradiated at ambient temperature. Data obtained in various fields. From Hahn et al. [1986].

pure Nb₃Sn, as shown, for example, in Figure 3.2.14. Hahn et al. [1986] reported neutron irradiation of Nb₃Sn superconductors with 0.5 and 1.5 wt.% Ti additions. The specimens were subjected to annealing at ~300 K following the irradiations at 5 K in the ANL IPNS Source. Additions of Ti to the binary system are of interest because of the increase produced in B_{c2} (§1.3.4). The specimens contained about 10 000 filaments of small size (~1.3 μm × 5 μm) and the bronze-to-Nb ratio was 3:1. Figure 3.3.1 from Hahn et al. [1986] presents the results as a function of damage energy, E_D, defined above, and Figure 3.3.2 from Weber [1986] presents the same data as a function of the fast neutron fluence (E > 0.1 MeV). Earlier data from ambient-temperature irradiations of the binary material from Snead and Parkin [1976] also presented in these figures show that a significant decrease in J_c of the newer, higher B_{c2} material occurs at a neutron fluence of about 10²²/m², much below the fluence of about 2 × 10²³/m² at which comparable J_c degradation is observed for the binary material. A comparison of Figure 3.3.1 with Figure 3.2.14 from the Söll review shows that the peak in J_c occurs at E_D ≈ 0.02 eV for the alloyed material as compared with E_D ≈ 0.2 eV for a number of binary Nb₃Sn systems tested by various investigators.

Subsequent irradiation at ambient temperature with 14-MeV neutrons from the LLNL RTNS source confirmed the greater radiation susceptibility of ternary, or alloyed Nb₃Sn compared to binary Nb₃Sn [Weiss et al., 1987]. Figure 3.3.3 shows results for both J_c and B_{c2} from two types of binary material: (A) a 10 000-core bronze-processed wire (reacted at 700°C for 105 h) and (B) a 19-core wire (reacted at 700°C for 64 h). The maximum J_c and B_{c2} appeared at a somewhat lower 14-MeV fluence of ~0.45 × 10²²/m² for sample B than the peak fluence of ~0.7 × 10²²/m² observed for sample A. The comparable maximums in both J_c and B_{c2} for Ti-, (Ni + Zn)-, and Ta-alloyed wires occurred at much lower fluences of about 0.2 × 10²²/m², and the enhancements of J_c were also smaller (Figure 3.3.4). Figure 2.3.1 in §2.3 showed that the depression in T_c for these alloyed specimens was only slightly larger than the T_c depression for the binary specimens. Weiss et al. suggested that the damage mechanism for binary Nb₃Sn was mostly homogeneous antisite disordering by replacement collisions whereas in alloyed materials such as Ta, Suenaga [citation, Weiss et al., 1987] had shown that the alloying enhanced the sensitivity of T_c to strain by a factor of two. Thus, by analogy, an additional mechanism

Figure 3.3.2. Normalized J_c vs. fast neutron fluence for an alloyed Nb_3Sn conductor irradiated at 5 K compared to binary Nb_3Sn irradiated at ambient temperature. Data obtained in various fields. From Weber [1986].

Figure 3.3.3. Normalized I_c and B_{c2} of two types of multifilamentary, binary Nb_3Sn after 14-MeV neutron irradiation at 25°C. From Weiss et al. [1987].

Figure 3.3.4. Normalized I_c and B_{c2} of three types of multifilamentary, ternary Nb_3Sn after 14-MeV neutron irradiation at 25°C. A-Ti-alloyed, B-(Ni+Zn)-alloyed, and C-Ta-alloyed. From Weiss et al. [1987].

connected with compressive strains should be invoked for the faster decrease of T_c with dose of the alloyed material.

Weiss et al. advanced the following explanation for the data in Figures 3.3.3 and 3.3.4. Following Equation (3.2.2), the increase of B_{c2} in the low fluence region ($<10^{22}/\text{m}^2$) was attributed to an increase in the normal state resistivity ρ_n . In this fluence region, T_c and γ , the electronic density of states, do not change appreciably. The increase in B_{c2} allows an increase in J_c that depends sensitively on the applied field. At 20 T, I_c/I_{c0} lies between 3 and 4, compared to $I_c/I_{c0} \approx 2$ at 14 T. The change in ρ_n has an exponential dependence on fluence, so the rate of change of I_c/I_{c0} and B_{c2} with fluence eventually decreases. At higher fluences, the degradation of T_c owing to atomic disordering can reach 20% or more (§2), and this effect will eventually lower J_c . For alloyed wires, the initial enhancement of ρ_n with irradiation is smaller than for binary wires, owing to the higher initial value of ρ_n for the alloyed wires. In binary wires, an enhancement of $J_c(B)$ in measurements in high fields after irradiation is correlated to the introduction of radiation-induced flux pinning centers as ρ_n increases, but in alloyed wires, this effect is reduced or even suppressed (Ta addition). However, Weiss et al. did not advance an explanation for the decrease in J_c below initial values in alloyed wires at relatively low irradiation fluences of about 1 to $2 \times 10^{22}/\text{m}^2$ ($E = 14$ MeV).

Further comparative investigations of commercial binary and ternary Nb_3Sn conductors are discussed below, in §3.4 on high energy neutron irradiation. The general conclusions that ternary material is more radiation-sensitive were confirmed (Figures 3.4.7 and 3.4.8).

3.4. HIGH ENERGY NEUTRON IRRADIATION

Comparative neutron irradiations at 14 MeV of alloyed and binary Nb_3Sn were discussed above [Weiss et al., 1987]. Earlier results by Scanlon and Raymond [1979] on two types of bronze processed binary material are presented in Figure 3.4.1. The specimens were irradiated near 4 K with high energy neutrons from two sources with the spectra shown in Figure 3.4.2. These measurements were made at 4 K in fields of 12 T on (A) laboratory monofilament

Figure 3.4.1. $\Delta I_c/I_{c0}$ vs. high energy neutron fluence for (A) Nb_3Sn monofilament and (B) multifilamentary Nb_3Sn in a field of 12 T at 4.2 K. Irradiation temperature was 4.2 K. From Scanlon and Raymond [1979].

Figure 3.4.2. Neutron spectra for the two neutron sources used in the irradiations for which data are shown in the preceding figure. From Scanlon and Raymond [1979].

Figure 3.4.3. I_c of 19-core Nb_3Sn vs. magnetic field after ambient-temperature, 14-MeV irradiation at several fluences. From Snead et al. [1981].

wires and (B) commercial multifilamentary samples (1045 filaments, 3- μ m diameter, bronze matrix). Perhaps the declines reported by Weiss et al. were not observed because the maximum irradiation fluences were below $\sim 6 \times 10^{21}/\text{m}^2$. At these fluences, no changes in T_c were observed within the experimental error of ± 0.5 K. As Figure 3.4.2 indicates, the different spectra of the two high energy sources imply different damage energies. Calculations predicted a factor of 1.5 for an equivalent increase in I_c after 14-MeV RTNS irradiation as compared to Be(d,n) irradiation. The results for the multifilamentary commercial sample scaled by exactly that amount, but the results on the laboratory monofilamentary specimens scaled by a factor of 1.7 to 2.5. The discrepancy may have been due to off-axis positioning of the laboratory specimen in the Be(d,n) source.

Snead et al. [1981] also carried out comparative irradiations of bronze processed Nb₃Sn with the same two sources for which the spectra are shown in Figure 3.4.2. However, these irradiations were made at ambient temperature. Results of 14-MeV RTNS irradiation of a 19-filament, laboratory specimen produced at BNL are shown in Figure 3.4.3. Decreases in I_c vs. the transverse field were observed, beginning at RTNS 14-MeV fluences of about $2 \times 10^{22}/\text{m}^2$. Results with the Be(d,n) source on single-core material are presented in Figure 3.4.4). Unfortunately, the peak fluence was lower than the highest 14-MeV fluence and the measurements were made on single-filament wires. Although comparative irradiations were also done in the HFBR, graphical or tabular results were not presented. However, a critical fluence was determined at which the change in J_c was zero in an applied field of 10 T. This fluence was $4.4 \times 10^{22}/\text{m}^2$ ($E > 0.11$ MeV) in the HFBR (corrected fluence compared with earlier publications of HFBR results) and $7.5 \pm 2.5 \times 10^{21}/\text{m}^2$ in the RTNS.

Results of 14-MeV neutron irradiation, but at 4 K, of a monofilament Nb₃Sn sample nearly identical to that of Snead et al. [1981] were presented by Guinan et al. [1984]. Temperature excursions to 61.5 K occurred during the cryogenic irradiation process, but the authors did not expect significant annealing to result. The single-filament sample was prepared by the bronze process method. Figure 3.4.5(A) gives I_c vs. field after each irradiation and Figure 3.4.5(B) compares these results at 10 T and 4 K with the earlier high-energy neutron results on very similar samples by Scanlon and Raymond [1970]

Figure 3.4.4. I_c of single-core Nb_3Sn vs. magnetic field after ambient-temperature, d-Be neutron irradiation at several fluences. From Snead et al. [1981].

Figure 3.4.5. (A) I_c vs. field of monofilamentary Nb_3Sn after each 14-MeV irradiation. Fluence units are $10^{22}/m^2$. (B) Normalized I_c vs. damage energy, E_D , for monofilamentary specimens. Closed symbols represent 4-K irradiation; open symbols represent ambient-temperature irradiation. From Guinan et al. [1984].

Figure 3.4.6. Normalized J_c in several types of Nb_3Sn conductors after 14-Mev and reactor irradiations vs. equivalent fluences (A). The relative number of neutrons/ m^2 which equal damage from 1 neutron/ m^2 in RTNS-II are also shown (B). From Kulcinski et al. [1986].

and by Snead et al. [1981]. One point from an ~4-K fission-spectrum irradiation at the ANL CP5 facility is also shown [Colucci and Weinstock, 1978] (see also Figure 3.2.5 above).

Wires prepared by the in situ process described above (§2.2.1) were also irradiated under the same conditions by Guinan et al. [1984]. The results, together with additional comparative measurements compiled by Kulcinski et al. [1986] are shown in Figure 3.4.6(A). A graph that shows the number of neutrons from both various radiation sources and earlier fusion designs that equal the damage from one RTNS neutron that is given as part (B) of this figure is based upon work by Guinan and coworkers [citations, Kulcinski et al., 1986]. Since the in situ processed wire does not exhibit a peak in J_c and appears more sensitive to radiation than the bronze-processed specimens, caution is indicated in applying results from the extensive data base on bronze material to composite conductors fabricated by other methods. Figures 3.4.6(A) and 3.4.5(B) both show that the 4-K, 14-MeV results on monofilament wire of Guinan et al. [1984] were not reproduced in any of the other investigations. However, Brown et al. [1975] also found higher initial increases in J_c after cryogenic irradiation than others found for similar material after ambient-temperature irradiations, although it is surprising that the equivalent reactor peak fluence of about $8 \times 10^{22}/\text{m}^2$ was not high enough to produce a decline in J_c (see Figure 3.2.9). Both Figures 3.3.2 and 3.3.3 indicate that the peak fluence is not strongly dependent upon magnetic field.

More recently, 14-MeV irradiations at the RTNS were carried out to establish a data base on commercial, multifilamentary Nb_3Sn [Hahn et al., 1991]. Both binary and alloyed conductors were tested; the characteristics of these conductors were listed above in Table 2.3.1. The wires were reacted according to the manufacturers' specifications for maximum J_c for $B > 12$ T. The TWC conductors may have been fabricated by the modified jelly roll process (§1.3.3) and the IGC conductors may have been fabricated by the internal tin process (§1.3.2), but this information was not reported. Most of the data reported pertain to ambient irradiations, but some of the irradiations were carried out at 14 K. Occasional temperature excursions from 14 K caused by sudden release of H_2 (from neutron damage to the electrical insulator,

Figure 3.4.7. Non-Cu J_c measurements at 15 T and 4.5 K for several commercial binary Nb₃Sn conductors irradiated at 14 to 25 K (LT) or at ambient temperature (RT) with 14-MeV neutrons. J_c has units of A/mm². See Table 2.1 for specifications. From Hahn et al. [1991].

polyimide) were limited to 25 K and a few seconds. The J_c measurements made at ~4.5 K in a 15-T field are presented in Figure 3.4.7 (binary) and Figure 3.4.8 (ternary Ti alloys). (Unfortunately, the quality of the graphics is below that which is customary for the journal in which these results were published.) In agreement with the results of Weiss et al. [1987] (Figures 3.3.3 and 3.3.4), the alloyed specimens showed a decline in J_c (below the initial value) at a lower fluence than that of the binary material. This critical fluence was about 0.8 to $1.3 \times 10^{22}/\text{m}^2$, or $E_D \approx 0.25$ to 0.35 . (The equivalent ITER TF-magnet fluence would be about 3 to $6 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV), based on the equation of a fission fluence ($E > 0.1$ MeV) to about 75% of the magnet fluence, and the fluence relationship shown in Figure 3.4.6(B) or the E_D -to-fluence relationship of Figures 3.3.1 and 3.3.2.) These results for Ti-alloyed specimens are in slight disagreement with some earlier fast neutron results: Hahn et al. [1986] and Weber [1986] found a steeper decline in J_c of about 50% at a fission fluence of $10^{22}/\text{m}^2$ ($E > 0.1$ MeV) or $E_D \approx 0.05$ eV, but their field strengths were lower. Weiss et al. [1986] measured J_c at 16 T and reported the peak to lie between 1 and $2 \times 10^{21}/\text{m}^2$ ($E = 14$ MeV). This is in good agreement with the results of Hahn et al. [1991] at 15 T. See Figures 3.3.1, 3.3.2, and 3.3.4.

Except for the in situ irradiated, binary TWG-247 conductor with poor initial J_c , the differences between low and ambient-temperature irradiations are within the uncertainties the authors expected from sample to sample. (Although the authors reported that six samples of each type of conductor were irradiated, they did not report the variability between samples.) Brown et al. [1978b] reported an irradiation-temperature effect for binary Nb_3Sn with a J_c of 2×10^4 A/mm² at 5 T, but no effect for material with a lower J_c of 8×10^3 A/mm² at 5 T (Figures 3.2.10 and 3.2.11, §3.2.2). An extrapolation of the J_c vs. B graph provided by Brown et al. indicates that the J_c of their best material could still be above 10^3 A/mm² at 15 T and 4 K. This appears to be significantly above the initial J_c values of the specimens of Hahn et al. [1991], which ranged from about 100 to 450 A/mm² at 15 T. Hence, the lower initial J_c values of Hahn et al. could explain their failure to observe differences between 4-K and ambient-temperature irradiation damage.

Figure 3.4.8. Non-Cu J_c measurements at 15 T and 4.5 K for several commercial ternary Nb₃Sn conductors irradiated at 14 to 25 K (LT) or at ambient temperature (RT) with 14-MeV neutrons. J_c has units of A/mm². See Table 2.1 for specifications. From Hahn et al. [1991].

For the binary specimens, the value of J_c at field initially increased and was above or only slightly below the initial value at the maximum fluence of about $2 \times 10^{22}/\text{m}^2$ ($E = 14$ MeV). However, the J_c of the alloyed conductors fell below the initial values at or slightly above a fluence of $1 \times 10^{22}/\text{m}^2$ ($E = 14$ MeV). This 14-MeV fluence is approximately equivalent to an ITER TF magnet fluence of $4 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV). (This equivalence is derived from Figure 3.4.6(B) and the correspondence of a reactor fluence to ~75% of a harder magnet fluence.)

3.5. EFFECTS OF STRAIN

A synergism between the effects of irradiation and strain on the J_c of Nb_3Sn was found by Fukumoto et al. [1985]. Reactor fast neutron fluences up to $4 \times 10^{23}/\text{m}^2$ ($E > 0.1$ MeV) were used at an ambient temperature estimated to be below 150°C . Material A was prepared with the "in situ" technique (see §2.2.1) whereas material B was formed by powder metallurgy. Specimen wires were 0.31 mm in diameter for material A and 0.20 mm for material B. Figure 3.5.1 shows the normalized J_c vs. intrinsic strain ϵ_0 ($= \epsilon - \epsilon_m$) diagrams for both materials. (As explained in §1.4, with or without irradiation, J_c increases with strain, ϵ , up to a maximum value at ϵ_m and then decreases when the material is strained above this value.) At the lower fluence of $2.8 \times 10^{22}/\text{m}^2$, the change in strain sensitivity of material A is within the difference exhibited by two unirradiated specimens of the material (also shown in the figure). However, the strain sensitivity is slightly higher in the specimens irradiated to $1.1 \times 10^{23}/\text{m}^2$ than in the other specimens. In material B, the strain sensitivity also appears to be enhanced by neutron irradiation, although the strain-induced decrease in J_c appears to be greater at the lowest fluence of $2.8 \times 10^{22}/\text{m}^2$. The highest fluence of $4 \times 10^{23}/\text{m}^2$ caused a decrease of J_c to ~0 in all materials. Although the measurements were made at a field of 6 T, a mathematical analysis by Fukumoto et al. showed that the synergism between the effects of irradiation and strain of J_c should be increased in higher fields.

Fukumoto's group continued measurements of radiation-enhanced strain sensitivity with higher strain measurement accuracy ($\pm 0.008\%$ strain) using bronze processed, fine multifilamentary wire [Okada et al., 1987]. The number

Figure 3.5.1. I_c normalized to maximum value vs. intrinsic strain before and after fast neutron irradiation at 60°C for Material A (in situ technique) and Material B (powder metallurgy). Nb_3Sn specimens. From Fukumoto et al. [1985].

of 5- μ m diameter filaments in the wire was 745 and the diameter of the conductor was 0.31 mm. The volume fraction of Cu stabilizer was 0.31, that of the Nb₃Sn was 0.16, that of Nb was 0.12, and that of the bronze was 0.41. The irradiation and measurement conditions were similar to those of the earlier work, but the irradiation temperature was estimated to be about 82°C and the effects of annealing on J_c were also investigated. Stress-strain curves at 4 K showing both the effects of irradiation and of annealing are given in Figure 3.5.2. Irradiation increases the yield stress and decreases the ductility, but annealing at 700°C can restore tensile characteristics. It is expected that hardening and embrittlement occur in the bronze matrix and the Cu stabilizer under irradiation. Figure 3.5.3 presents the effect of strain on J_c before (A) and after (B) irradiation to a fast neutron fluence of $1.5 \times 10^{23}/\text{m}^2$ ($E > 0.1$ MeV) in fields up to 6 T. The value of ϵ_m is increased, the intrinsic strain at irreversibility, $\epsilon_{0,\text{irrev}} = \epsilon_{\text{irrev}} - \epsilon_m$, decreases, and the strain at failure, ϵ_f , decreases. Figure 3.5.4 shows the effects on these defined strains for other fluences. The marked increase in ϵ_m with fluence was thought to imply that additional compressive strain in the direction of the axis occurred to the Nb₃Sn filaments during the irradiation through an unknown mechanism. It was expected that this increase in ϵ_m would also occur even if the conductor were irradiated at cryogenic temperature.

Since the addition of Ti improves the strain sensitivity of Nb₃Sn conductors [citation, Katagiri et al., 1993], measurement of synergistic irradiation-strain effects in (Nb,Ti)₃Sn is of interest. Katagiri et al. [1993] recently measured changes in ϵ_m , ϵ_{irrev} , and ϵ_f (defined above) for commercial, bronze processed (Nb,Ti)₃Sn multifilamentary wires. The ambient-temperature radiation resistance of the ternary material appeared to be more like that of the binary system and better than the cryogenic radiation resistance of the ternary material tested by Weber [1986] and Hahn et al. [1986], indicated by the dashed line in Figure 3.5.5 (see also Figures 3.3.1 and 3.3.2 above). The amount of Ti, 1.8 at.%, was significantly less than that of Weber (2.6 at.%), but also, the specimens measured by Weber and Hahn et al. were irradiated at ~5 K. The changes in the strain sensitivity parameters of the ternary material are shown in Figure 3.5.6. Again, ϵ_m increased with irradiation fluence, as it had for the binary material (dashed line in Figure 3.5.6). The ϵ_{irrev} initially decreased slightly and then recovered. However, there is a

Figure 3.5.2. Stress vs. strain curves at 4 K before and after irradiation and annealing for bronze processed, Nb_3Sn multifilamentary composite wire. Irradiation was performed at $<355 \text{ K}$. From Okada et al. [1987].

Figure 3.5.3. I_c vs. strain (A) before irradiation and (B) after irradiation to a fast neutron fluence of $1.5 \times 10^{23}/m^2$ ($E > 0.1$ MeV) at various fields. Specimens were multifilamentary Nb_3Sn composite wire. From Okada et al. [1987].

Figure 3.5.4 Strain vs. fast neutron fluence on the characteristic strains defined in the text. From Okada et al. [1987].

Figure 3.5.5. Normalized I_c at two strains vs. fast neutron fluence ($E > 0.1\text{ MeV}$) for several superconducting wires. From Katagiri et al. [1993].

Figure 3.5.6. Changes in characteristic strain values (defined in text) vs. neutron fluence for ternary Nb_3Sn wires. From Katagiri et al. [1993].

larger rise in ϵ_{irrev} at a fluence of $3 \times 10^{23}/\text{m}^2$ ($E > 0.1$ MeV) in the ternary material than in the binary material. The decrease of ϵ_f with fluence and its eventual coincidence with ϵ_{irrev} is similar in both ternary and binary material. Katagiri et al. reported that the increase in ϵ_m was not fully explained by the hardening of ductile materials in the conductor. The coincidence of ϵ_f and ϵ_{irrev} was explained as a hardening and embrittlement of the bronze matrix by high-fluence irradiation, which does not allow crack blunting of the bronze surrounding the Nb_3Sn filament. The ϵ_{irrev} increases owing to the enhancement of ϵ_m with irradiation and so does ϵ_f . The decrease in $\epsilon_{0,\text{irrev}}$ ($= \epsilon_{\text{irrev}} - \epsilon_m$) with increasing fluence was attributed to embrittlement of the filament, but was less significant than for the binary material, perhaps because the irradiation-induced suppression of the martensitic transformation was less significant with the Ti addition.

When the irradiation fluence is expressed in terms of damage energy, E_D , in units of eV/atom, where $4 \times 10^{22}/\text{m}^2 = 1$ eV/atom ($E = 14$ MeV), then $\epsilon(E_D) \approx [\Delta c/c](E_D) = 0.00165/\Delta T_c(E_D)$, where c is the lattice parameter [Summers et al., 1991]. This expression for ϵ is valid for small changes in T_c (less than 6 to 8 K). The effect of irradiation on T_c in terms of E_D was discussed above, in §2.5. The radiation-induced strain $\epsilon(E_D)$ in this expression evidently refers to the intrinsic strain, ϵ_0 , since Summers et al. use ϵ for Ekin's ϵ_0 when they quote his equations (see also §1.4). Thus, this result cannot be directly compared with the data of Katagiri et al. [1993].

Figure 4.1. B_{c2} vs. temperature for $(\text{Nb,Ta})_3\text{Sn}$. From Hampshire et al. [1985].

4. RADIATION-INDUCED CHANGES IN THE UPPER CRITICAL FIELD

There have been fewer postirradiation measurements of the upper critical field, B_{c2} , than of T_c and J_c . B_{c2} is defined as the applied field at which the magnetization of a type II superconductor in the mixed state goes to zero, but for practical purposes, B_{c2} is sometimes defined as the transverse field at which J_c falls to 0.1 A/mm^2 and is determined by extrapolation in a J_c vs. B plot [citations, Coffey et al., 1967]. (When values of the field are given in units of T, rather than A/m, the label on the graphs reproduced here should be B_{c2} , not H_{c2} .) As Figure 4.1. indicates, B_{c2} is a sensitive function of temperature. This fact is important when irradiation results at different temperatures are compared.

4.1. BULK, BINARY Nb_3Sn

4.1.1. Ambient-Temperature Irradiation

Large fractional reductions in B_{c2} of vapor-deposited Nb_3Sn were reported by Sweedler et al. [1975] after neutron irradiation at 60°C in the HFBR. For unirradiated Nb_3Sn , B_{c2} at 4.2 K was 23.5 T; after a fluence of $5.0 \times 10^{23}/\text{m}^2$ ($E > 1 \text{ MeV}$), B_{c2} at 1.6 K was 0.03 T. Karkin et al. [1976] also observed significant decreases in B_{c2} after ambient-temperature neutron irradiation (E unspecified). The fluence dependence is shown in Figure 4.1.1, together with a single measurement obtained by Schweitzer and Parkin [1974]. The values of $B_{c2}(0)$ were determined from the temperature dependence of $B_{c2}(T)$ obtained within the temperature interval from 2 to 18 K and an analytic expression given by Karkin et al. The observation of a significant decline in B_{c2} at a fluence of $5 \times 10^{23}/\text{m}^2$ is in agreement with the results of Sweedler et al. [1975] discussed above. Annealing to about 800°C was required to restore B_{c2} to its original value (Figure 4.1.2). These specimens were produced by baking pressed pellets of Nb and Sn powder at 1050°C for 20 h; Figures 2.1.5 and 2.4.2 show results of T_c measurements on these specimens.

Figure 4.1.1. $H_{c2}(0 \text{ T})$ vs. fast neutron fluence after ambient-temperature Nb_3Sn irradiation. Δ —data from Schweitzer and Parkin [1974]. Figure from Karkin et al. [1976].

Figure 4.1.2. $H_{c2}(0 \text{ T})$ vs. isochronal annealing temperature for Nb_3Sn specimens irradiated to fast neutron fluences of $F \times 10^{23}/\text{m}^2$. Δ - $F = 4$. $\circ$ - $F \approx 7$. From Karkin et al. [1976].

Figure 4.1.3. $H_{c2}(0 T)$ of Nb_3Sn vs. T_c depressed by irradiation. The arrows represent the increase in H_{c2} values during the isochronal annealing; the end points represent the values after 300-K recovery. From Besslein and Ischenko [1976].

4.1.2. Cryogenic-Temperature Irradiation

Coffey et al. [1967] measured B_{c2} of vapor-deposited Nb_3Sn before and after 15-MeV deuteron irradiation at about 25 K. After a deuteron fluence of $10^{21}/m^2$, B_{c2} decreased from 4.4 to 2.3 T at measurement temperatures of 0.810 T_c and 0.883 T_c , respectively. (B_{c2} is a sensitive function of the measurement temperature, as Figure 4.1 indicates, so fractional reductions in B_{c2} were lower than those of Sweedler et al. quoted above, because the measurements were made at higher temperature.) At similar, relatively high temperatures of measurement, Coffey et al. found that B_{c2} increased to 2.42 T after a 77-K anneal and to 2.72 T after a 300-K anneal.

Besslein and Ischenko [1976] found that a monotonic decrease in B_{c2} almost proportional to the decrease in T_c occurred after heavy ion irradiation below 30 K of layer specimens of Nb_3Sn . These specimens were 5 μm thick. Figure 4.1.3 shows that the decrease in B_{c2} followed the decrease in T_c with increasing irradiation fluence. (The fluence increases from right to left in the figure.) The expression $dB_{c2}/dT_c = 0.95$ T/K was derived [Ischenko et al., 1978].

4.2. COMPOSITE, BINARY Nb_3Sn

4.2.1. Ambient-Temperature Irradiation

Neutron irradiation at the HFBR at BNL at $\sim 80^\circ C$ on multifilamentary specimens showed a relationship between B_{c2} and T_c similar to that found by Besslein and Ischenko on bulk material [Ostenson et al., 1983]. Figure 4.2.1 indicates that the fluence dependence of both B_{c2} and T_c to $1.3 \times 10^{23}/m^2$ ($E > 1$ MeV) was comparable. These data were obtained on specimens prepared by an in situ casting and drawing technique that produces thin (~ 70 -nm thick) superconducting filaments that are discontinuous (§2.2.1).

4.2.2. Cryogenic-Temperature Irradiation

Maier and Seibt [1981] also found that the relationship between B_{c2} and T_c after irradiation was nearly linear, as Figure 4.2.2 indicates. These

Figure 4.2.1. Comparison of the dependence of H_{c2} and T_c of in situ processed Nb_3Sn upon fast neutron fluence ($E > 1$ MeV) after irradiation at $-80^\circ C$. From Ostenson et al. [1983].

Figure 4.2.2. T_c vs. B_{c2} for bronze processed Nb_3Sn superconductors with different reaction temperatures T_R . Curves a and b are theoretical. From Maier and Seibt [1981].

Figure 4.2.3. H_{c2} for an Nb₃Sn monofilament wire vs. temperature before and after ~6-K, fast neutron irradiation and after an anneal. From Colucci and Weinstock [1978].

irradiation investigations were carried out with 50-MeV deuterons at 18 K up to a maximum fluence of $2.3 \times 10^{22}/\text{m}^2$. The Nb_3Sn bronze-processed single- and multicore specimens had different layer thicknesses at the five reaction temperatures indicated in the figure.

Colucci and Weinstock [1978] irradiated filamentary Nb_3Sn reacted by the bronze process at 700°C for 24 h. Their type B material contained one filament of $5.7 \times 10^{-6}\text{cm}^2$ cross-sectional area and had $J_c = 1.6 \times 10^4\text{A}/\text{mm}^2$ at 4 T and 4.2 K before irradiation. The irradiation was carried out at the CP-5 reactor at ANL at a temperature of ~ 6 K. Figure 4.2.3 shows the dependence of B_{c2} on temperature before and after a relatively low fluence irradiation to $1.8 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV). As expected, the decrease in B_{c2} is less pronounced than that after a fluence $> 10^{23}/\text{m}^2$. Annealing to 540 K restored B_{c2} at the lower measurement temperatures.

4.3. COMPOSITE, TERNARY Nb_3Sb

In addition to the relationship between damage to T_c and to B_{c2} , a correlation between the irradiation changes in J_c and B_{c2} has also been demonstrated, for ternary as well as binary composite Nb_3Sn . This relationship was shown above, in Figures 3.3.3 and 3.3.4, which indicate that peak values of both parameters occurred at similar irradiation fluences after ambient 14-MeV neutron irradiation [Weiss et al., 1987]. Hahn et al. [1986] cited data from Snead that showed that the maximum enhancement of B_{c2} (about 12%) occurred at the same fluence as the peak in J_c . The increase in J_c with irradiation was thought to be caused by the increase of B_{c2} via the radiation-induced increase of ρ_n [Equations (3.2.1) and (3.2.2)]. Hahn et al., who studied the effect of fast neutron irradiation at 5 K on ternary Nb_3Sn compounds $[(\text{Nb},\text{Ti})_3\text{Sn}]$ stated that if the increase in B_{c2} in the unirradiated ternary superconductor was caused at least partly by the same mechanism, (i.e., preirradiation disorder introduced by the alloying of the third component), then an almost immediate decrease in J_c with neutron fluence would be expected in ternary compounds. This was observed, as Figure 3.3.2 above indicates. This conclusion was also supported by the observation that for a given Ti concentration, the J_c degraded faster in the material with the higher preirradiation value of B_{c2} .

Table 4.3.1. B_{c2} Estimates at 4.5 K Based on Ambient-Temperature Irradiation.
Fluences in Units of $10^{22}/\text{m}^2$ ($E = 14$ MeV). From Hahn et al. [1991].

SEWC		SUMI		TWC-247		TWC-246		TWC-325	
ϕ_t	B_{c2} (T)	ϕ_t	B_{c2} (T)	ϕ_t	B_{c2} (T)	ϕ_t	B_{c2} (T)	ϕ_t	B_{c2} (T)
0.170	20.02	0.000	20.74	0.170	18.69	0.000	21.67	0.000	21.64
0.427	19.97	0.170	21.07	0.427	19.24	0.427	21.89	0.170	21.94
0.563	20.33	0.427	23.20	0.545	19.31	0.773	22.46	0.427	21.88
0.843	19.65	0.819	23.73	0.795	19.48	1.495	19.84	0.713	21.81
1.824	19.01	1.686	23.19	2.005	18.97	2.009	18.88	1.359	19.43
2.517	81.78*	2.261	22.44					2.017	18.90

* Probably a typographic error for 18.78 T.

Table 4.3.1. presents additional B_{c2} data on both binary and ternary specimens (TWC-246 and TWC-325) after ambient-temperature irradiation with RTNS 14-MeV neutrons [Hahn et al., 1991]. The characteristics of these commercial, multifilamentary specimens were given above, in Table 2.3.1. The J_c data for these specimens were shown above, in Figures 3.4.7 and 3.4.8. In general, the results given in Table 4.3.1 are in accord with those of Weiss et al. [1987] shown in Figures 3.3.3 and 3.3.4. The ternary specimen with the higher Ti content (TWC-325) exhibited a somewhat less pronounced peak in B_{c2} with fluence. However, at the maximum fluences of about $2 \times 10^{22}/\text{m}^2$ ($E = 14$ MeV), all specimens, binary and ternary, exhibited a similar B_{c2} of nearly 19 T (at 4.5 K), except for the SUMI specimen, which had a B_{c2} of 22.4 T.

From analysis of the data of Hahn et al. [1991], Summers et al. [1991] expressed the change in T_c as a function of damage energy, E_D :

$$T_{c0m}(E_D)/T_{c0m}(0) = \exp(-E_D/4), \quad (4.3.1)$$

where E_D is in eV/atom and for 14-MeV neutrons, $1 \text{ eV/atom} \approx 4 \times 10^{22}/\text{m}^2$. Then they derived the corresponding change in B_{c2} as

$$B_{c20m}(\epsilon, E_D)/B_{c20m}(0) = [(\rho_n + 600E_D)/\rho_n] \cdot [\exp(-5E_D/8)] \cdot [1 - a|\epsilon_M + \epsilon(E_D)|^u], \quad (4.3.2)$$

where ρ_n is the normal state resistivity of Nb_3Sn (without radiation), $u = 1.7$, and $a \approx 900$ for $\epsilon < 0$ and $a \approx 1250$ for $\epsilon > 0$. The quantities B_{c20m} and T_{c0m} are the values of B_{c20} and T_{c0} at zero intrinsic strain. The best fit was obtained with $B_{c20m} = 28.7$ T and $T_{c0m} = 18.3$ K. The mechanical strain is ϵ_M and the irradiation strain is $\epsilon(E_D)$. The definition of ϵ corresponds to that of ϵ_0 as given in §1.4. A comparison of calculated values from this equation with experimental data had not been published at this writing.

Figure 5.1.1. The Al₅ structure: A₃B. From Fähnle [1981].

5. X-RAY STUDIES OF THE ORDER PARAMETER AND LATTICE EXPANSION

Attempts to explain the radiation sensitivity of Nb₃Sn in terms of fundamental properties are of interest to this review, because if such attempts are successful, the data base is strengthened.

5.1. ORDER PARAMETER

Numerous attempts have been made to correlate the observed decreases with radiation of T_c , J_c , and B_{c2} with the order parameter, S . The A15 structure consists of a bcc B atom sublattice and three non-intersecting orthogonal linear A atom chains, as shown in Figure 5.1.1 from Föhnle [1981]. The original antisite defect model assumed that the main effect of irradiation was the replacement of some A (Nb) atoms on the linear chain with B (Sn) atoms. This disordering can be expressed by the Bragg-Williams long-range order parameter

$$S_A = (P_A - r)/(1 - r) \quad (5.1.1)$$

where $0 \leq S \leq 1$, P_A is the probability of an A site being occupied by an A atom, and r is the fraction of A atoms in the binary alloy. If all A sites are occupied by A atoms, $S_A = 1$.

In the earliest studies of the relationship with S of the superconducting properties T_c , J_c , or B_{c2} after irradiation, S was not measured independently. Instead, an analytic expression relating S to the irradiation fluence was used, and not unexpectedly, a correlation between the superconducting properties and the derived S value was exhibited. Examples of such studies are those of Besslein et al. [1975], Sweedler et al. [1974], and Söll et al. [1976]. A slightly different approach was used by Brown et al. [1978a], who used a definition of S based on ρ_n , which again reflects lattice damage.

Sweedler et al. [1978] made fluence-independent and detailed order parameter measurements on irradiated A15 superconductors with X-ray and neutron diffraction, but studied primarily Nb₃Al and Nb₃Ge. They found clear indications of decreasing order with successive irradiations and a correlation of the decrease in T_c with a decrease in S_A .

A

B

Figure 5.1.2. (A) X-ray diffraction peaks 210 and 211 of an Nb₃Sn diffusion layer before and after fission fragment irradiation to a fluence of $2 \times 10^{19}/\text{m}^2$ at ambient temperature. The diamond diffraction peak is used as a standard. (B) Recovery of the 211 diffraction-line halfwidth, B , lattice parameter a , and the degree of long range order, S , during isochronal 15-min anneals after irradiation. From Skvortsov et al. [1978].

Skvortsov et al. [1978] used X-ray diffraction to measure S of Nb_3Sn diffusion tapes prepared by liquid Sn diffusion into an Nb substrate at 950°C . The thickness of the diffusion layer attained was more than $6\text{ }\mu\text{m}$. The maximum fission fragment irradiation fluence of $2 \times 10^{19}/\text{m}^2$ corresponded to a fast neutron fluence of about $10^{26}/\text{m}^2$ ($E > 1\text{ MeV}$) in terms of the energy available for elastic collisions. At this high fluence, complete disordering was observed ($\Delta S = 1$), but the Nb_3Sn retained the A15 structure despite the likelihood that each atom was displaced several times from its lattice site. The changes in S were deduced from the changes in the (211):(210) diffraction peak intensity ratios shown in Figure 5.1.2(A). Figure 5.1.2(B) shows the recovery of the diffraction-line halfwidth B , the lattice parameter a , and S during subsequent 15-min isochronal anneals at the indicated temperatures. In accord with post-annealing measurements of T_c and J_c , recovery of S occurred at about 800°C . As noted above, the fluence was large enough that each atom should have been displaced, yet the A15 structure was retained. This retention of structure was said to preclude the possibility of formation of amorphous regions in the Nb_3Sn material.

Burbank et al. [1979] performed detailed X-ray determinations of the crystal structure of Nb_3Sn films as a function of 1.9-MeV, ^4He -particle radiation damage. The consecutive irradiation fluence of annealing temperature and the x parameter, a measure of antisite defects, are shown in Table 5.1 to be initially correlated with T_c . The results through the first anneal at 800°C were as expected, with an increase in antisite defects as the total irradiation fluence increased, followed by a reduction in defects after the anneal. However, the subsequent annealing and irradiation results were not understood by the authors. They explained that the 200-nm (2000-Å) thick films that they studied posed problems for X-ray diffraction studies, owing to their highly developed preferred orientation. Also, a reduction in intensity following the second anneal suggested a loss of ~10% of the material from the particular set of crystallites included in the measurement set. Formation of a second phase, crystal reorientation, and grain boundary growth are possible explanations for the puzzling results after the second anneal.

From additional results, these investigators concluded that the displacements of both Nb and Sn atoms from their positions were rather large, greater than

Table. 5.1. Antisite Disorder in Nb₃Sn Films. From Burbank et al. [1979].

Fluence/Temperature, Time	T _c , K	x (Antisite)
Unirradiated	18.1	--
4.0 × 10 ²⁰ /m ²	10	0.208
4.0 × 10 ²⁰ /m ²	4.0	0.458
7.6 × 10 ²⁰ /m ²	2.95	0.483
800°C, 1 h	17.7	0.158
900°C, 2 h	17.7 +	0.308
900°C, 12 h	17.7 +	0.308
1 × 10 ²⁰ /m ²	--	0.308

0.02 nm (0.2 Å). They believed that the large displacements were responsible for the reduction of T_c . Burbank et al. also stated that they did not know of another class of materials in which the lattice integrity was maintained despite the release of virtually all atoms from their equilibrium sites. This behavior was ascribed to the A15 bonding. As described above, Skvortsov et al. also observed the preservation of the A15 structure despite considerable displacement damage. Burbank et al. suggested that the atoms were rocking off their equilibrium positions and that the Nb chains were expanding, with the nature of the bonding between Nb atoms changing from covalent to metallic. (The Nb-Nb distance in metallic Nb is 0.02 nm (0.2 Å) larger than the corresponding distance in covalently bonded Nb.) Lattice expansion data for Nb_3Sn are given in §5.2 below.

Another theory of radiation damage in Nb_3Sn proposed by Pande [1978; citations therein] was that small regions of very low T_c were introduced into a matrix retaining essentially the original T_c of the material. The composite system would then have a well-defined, but lower T_c due to the "proximity effect" of the regions of low T_c . Jenkins et al. [1982] used TEM (transmission electron microscopy) to search for disordered zones within cascade damage regions after bombardment of Nb_3Sn specimens by 20- to 60-keV Au^{++} ions at ambient temperature. The specimens were formed by diffusion of molten Sn into Nb disks at 930 to 960°C. After irradiation, a strain field that could arise from a relative volume misfit of ~1.5% was found. Jenkins et al. thought that antisite exchange was the most likely explanation for these results. They also found that damage was produced heterogeneously at cascade sites, and that a non-uniform model, such as that of Pande, would be appropriate to describe the effects of radiation on superconducting properties. But because they also found that antisite disorder was likely, they suggested that a combination of both theoretical models was appropriate.

Nölscher and Saemann-Ischenko [1985] also used X-ray diffraction to measure the decrease in T_c with decrease in S_A . These results are presented in Figure 5.1.3. (Hydrogenation was used to vary superconducting and transport properties over a wide range in the film specimens.) The irradiation was carried out below 25 K using 20-MeV S ions.

Figure 5.1.3. T_c vs. integrity of Nb chains. r_A , and the order parameter, S_A for differently damaged Nb_3Sn layers. The parameter r_A corresponds to P_A in Equation (5.1). From Nölscher and Saemann-Ischenko, 1985].

5.2. LATTICE EXPANSION

Bauer et al. [1975] found irradiation-induced lattice expansions up to 0.5% in Nb₃Sn wires that were irradiated at ambient temperatures with several different particles. Their results for T_c and J_c were presented above in Figures 2.1.2 and 3.1.4. Their plot of the observed decrease in T_c vs. the lattice expansion is shown in Figure 5.2.1.

Nölscher and Saemann-Ischenko [1985] also plotted the variations of T_c under irradiation against the lattice parameter, a_0 , as Figure 5.2.2 indicates. (Hydrogenation was employed to vary superconducting and transport properties over a wide range in the film specimens.)

Skvortsov et al. [1978] observed a lattice expansion of 1.25%, apparently the highest recorded. At ambient temperature, the lattice parameter increased from 0.5291 to 0.5357 nm (5.291 to 5.357 Å) and at 77 K, the lattice parameter increased from 0.5279 to 0.5345 nm (5.279 to 5.345 Å). The recovery of the lattice parameter was shown above, in Figure 5.1.2 (B). This expansion of 1.25% resulted from fission fragment irradiation at a temperature below 40°C to a fluence of $2 \times 10^{19}/\text{m}^2$. This fission fragment fluence was said to correspond to a neutron fluence of about $10^{26}/\text{m}^2$ ($E > 1$ MeV).

Figure 5.2.1.1. Decrease in T_c vs. reduced lattice expansion for several irradiations of Nb₃Sn wires. From Bauer et al. [1975].

Figure 5.2.2. T_c vs. lattice parameter at ambient temperature of Nb_3Sn films irradiated below 25 K with S ions. Data from hydrogenated and annealed specimens are also shown. From Nölscher and Saemann-Ischenko, 1985].

6. CONCLUSIONS

6.1. RADIATION LIMITS FROM TRANSITION-TEMPERATURE MEASUREMENTS

A series of measurements of T_c on bulk Nb_3Sn after ambient-temperature irradiation indicated that T_c was seriously degraded to half of its pre-irradiation value at a fast neutron fluence of about $4 \times 10^{23}/m^2$ ($E > 0.1$ MeV) [Figure 2.1.4]. At about $1.3 \times 10^{23}/m^2$, however, only about a 20% decrease in T_c was observed. A review of early data by Söll et al. [1976] (Figures 2.1.8 and 2.1.9) indicated that cryogenic irradiation results were similar to ambient-temperature irradiation results on bulk Nb_3Sn . However, T_c values for binary Nb_3Sn composite specimens prepared by various methods diverged from, and sometimes, fell well below the line established for ambient-temperature irradiation of bulk Nb_3Sn , as Figure 2.2.2 from Kodaka et al. [1985] indicates.

Cryogenic, 14-MeV irradiation of bronze processed, single-filament Nb_3Sn indicated that only about a 1.5-K decrease in T_c should be expected for a fluence of $1.3 \times 10^{22}/m^2$. This fluence could be equated to a reactor fast neutron fluence of $\sim 7 \times 10^{22}/m^2$ ($E > 0.1$ MeV) [e.g., Kulcinski et al. [1986], Figure 3.4.6 (B)]. Reasonable agreement for the change in T_c of this monofilamentary material after ambient and cryogenic irradiation from various sources was found when the data were plotted as a function of damage energy, E_D (Figure 2.2.4, Guinan et al. [1984]).

For ternary material, two groups that used 14-MeV neutron irradiation reported a more pronounced decrease in T_c of about 0.5 to 1 K compared to the decrease in the binary material at a given fluence. See Figures 2.3.1 [Weiss et al., 1987] and Figures 2.3.2 and 2.3.3 [Hahn et al., 1991]. The T_c of $(Nb,Ti)_3Sn$ decreased from about 17 to 14 K after a 14-MeV fluence of ~ 2.5 to $3 \times 10^{22}/m^2$. The ITER TF magnet fluence equivalent to a 14-MeV fluence of $2.5 \times 10^{22}/m^2$ is about $1.1 \times 10^{23}/m^2$ ($E > 0.1$ MeV), according to the fission factor given in Figure 3.4.6(B) and a proration of this fluence by 0.75 for a harder spectrum.

There is very little information on the change in T_c of ternary materials after cryogenic irradiation. Data are limited to a few points in Figure 2.3.3

on a ternary alloy from one manufacturer, IGC. These specimens were probably fabricated by the internal Sn method.

6.2. RADIATION LIMITS FROM CRITICAL-CURRENT MEASUREMENTS

6.2.1. Binary Nb₃Sn

6.2.1.1. Fast Neutron Irradiation

Results for ambient-temperature irradiations of bulk Nb₃Sn specimens indicated that an initial increase in J_c with irradiation was often observed, especially in specimens with low preirradiation values of J_c . At a fast neutron fluence of about $2 \times 10^{23}/\text{m}^2$ ($E > 0.1$ MeV), however, substantial degradation of J_c (except above 6 T) occurred in specimens that had initially experienced an enhancement (Figure 3.1.4). In ambient-temperature irradiation of composite, bronze processed Nb₃Sn, Parkin and Sweedler [1975] found no initial J_c increase when measurements were made in fields up to 4 T, but Snead and Parkin [1976] found initial J_c increases in higher fields of ~10 to 12 T. These enhancement effects had leveled off at a fast neutron fluence of $\sim 5 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV) and degradation of J_c began at or below $10^{23}/\text{m}^2$ and was nearly complete by $\sim 3 \times 10^{23}/\text{m}^2$ (Figure 3.3.2). Cryogenic irradiation results on similar specimens showed J_c enhancement even at the lower fields of about 3 or 4 T, presumably because of greater retention at low temperature of flux-pinning defects that would increase J_c . However, after cryogenic irradiation, a decrease in J_c to below the initial value (degradation point) occurred at $\sim 3 \times 10^{22}/\text{m}^2$ (Figure 3.2.9 [Brown et al., 1977a]). The energy range of the fluence was not specified and this equivalent fast neutron fluence was obtained by calibration of the T_c change in the specimen with results from another investigation. When Söll [1978] reviewed a number of early investigations on binary Nb₃Sn and normalized them to $J_{c\text{Max}}$ and to an equivalent fast neutron fluence (Figure 3.2.14), a decrease in J_c below the initial value (degradation point) occurred at approximately 4 to $8 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV) or at an average damage energy, E_D of 0.5 to 1.2 eV. The review of Söll included some cryogenic irradiations.

6.2.1.2. Conversion of 14-MeV Neutron Irradiation Data

After 14-MeV irradiation, Hahn et al. [1991] found that the degradation point for J_c occurred at about 2 to $3 \times 10^{22}/\text{m}^2$ ($E = 14$ MeV) [$E_D > \sim 0.6$ eV] when several types of commercial, binary Nb_3Sn composites were tested after both ambient and cryogenic irradiation (Figure 3.4.7). Use of the ratios shown in Figure 3.4.6(B) gives equivalent fast neutron reactor fluences of 1.1 to $1.7 \times 10^{23}/\text{m}^2$ ($E > 0.11$ MeV) for the drop of J_c below the initial value. Although calculated E_D values are similar, these degradation fluences are considerably above those of preceding investigations on binary Nb_3Sn summarized by Söll, so the validity of these conversion ratios is questionable for J_c damage in Nb_3Sn (unless the reported 14-MeV fluence values are incorrect, i.e., too high).

Kulcinski et al. [1986] compiled several other high energy and reactor-neutron investigations that demonstrated that the J_c of binary composites dropped below its initial value at $\sim 8 \times 10^{22}/\text{m}^2$ ($E > 0.11$ MeV) [Figure 3.4.6(A)]. These results are in better agreement with those in Söll's review, but the 14-MeV, 4-K irradiation of a monofilament specimen was not brought into correspondence with reactor fast neutron irradiation of the same material by the equivalence relations, and again, did not seem to agree with the earlier literature on cryogenic irradiation. See, for example, Figure 3.2.9 from Brown et al. [1977a]. Again, either the reported 14-MeV fluences are too high, or the conversion ratios are not valid, or there is something unique about the material, in that its degradation point occurs at an unusually high fluence.

6.2.2. Ternary Nb_3Sn

6.2.2.1. Comparison of Fast Neutron and 14-MeV Data

The use of ternary conductors is probably necessary to achieve the performance required in ITER (Table 1.3.1). However, when the newer ternary or alloyed Nb_3Sn composites were tested, degradation of J_c occurred at fluences below those required for binary Nb_3Sn . Using equivalent fast neutron fluences ($E > 0.1$ MeV) for 14-MeV neutron irradiation indicated that for Ti-alloyed Nb_3Sn ,

degradation of J_c below the initial value might occur above $3.3 \times 10^{21}/\text{m}^2$ (Figure 3.3.2) or $6 \times 10^{22}/\text{m}^2$ (Figure 3.4.8) or $17 \times 10^{22}/\text{m}^2$ (Figure 3.3.4(A), field = 16 T). The last two values are derived from primarily ambient-temperature, 14-MeV irradiations ($0.8 < \text{Ti} < 2.0 \text{ wt.}\%$), whereas the first, lowest degradation fluence is the direct result of cryogenic fast neutron irradiation of a 1.5 wt.% Ti specimen in fields of 4 or 8 T in the ANL facility. The 14-MeV fluences were equated to reactor fast neutron fluences with $E > 0.11 \text{ MeV}$ using the factors in Figure 3.4.6(B), but the lack of agreement with the directly-obtained fast neutron data casts doubt either on this procedure or on the reported 14-MeV fluences, or on the use of ambient-temperature irradiation data to predict cryogenic performance.

Although there is good agreement in the data from various investigators on binary, bronze processed Nb_3Sn , there is a lack of cryogenic irradiation data in the literature on ternary Nb_3Sn conductors fabricated by the newer procedures such as internal tin and modified jelly roll and a lack of agreement in the small amount of existing data. Some of these newer materials are currently in the procurement process for ITER prototype coils (Table 1.3.1). The 14-MeV results of Hahn et al. [1991] on ternary Nb_3Sn were obtained on material with initial J_c values below the current ITER procurement specifications as given in Table 1.3.1, even allowing for a decrease in the field from 15 to 12 T. Figure 3.4.8 shows that the initial J_c values ranged from about 370 to 450 A/mm^2 (15 T), whereas specifications require 550 and 700 A/mm^2 (12 T). Apparently, none of the newer candidate conductors under procurement that have adequate, higher J_c were tested for radiation sensitivity by Hahn et al. [1991]. Unless the TF magnet fluences remain below 75% of $3.3 \times 10^{21}/\text{m}^2$ ($E > 0.1 \text{ MeV}$), it would be prudent to undertake additional cryogenic irradiation testing of these new conductor types. A recent review by Söll [1993] also suggested further cryogenic irradiations. With the available, rather sketchy published cryogenic information on ternary materials limited to the few points shown in Figures 3.3.2 and 3.4.8, it appears unwise to increase ITER fast neutron fluences above $2.5 \times 10^{21}/\text{m}^2$ ($E > 0.1 \text{ MeV}$, magnet spectrum) without further cryogenic testing. This value represents the worst-case result for onset of deterioration of J_c [Weber, 1986]. This result was obtained on a modified-jelly roll-processed, 1.5-wt.% Ti ternary specimen

with J_c probably of the order of 1000 A/mm² at 12 T. (See Smathers et al. [1986] for data on similar laboratory specimens.)

The measurement of J_c on any type of Nb₃Sn materials after cryogenic irradiation with high fast neutron fluences is extremely limited. Brown et al. [1977a] used ²³⁵U doping to extend their cryogenic fluence above $2 \times 10^{22}/\text{m}^2$ (Figure 3.2.9). Hahn et al. [1991] irradiated three types of specimens at 14 K (excursions to 25 K) with 14-MeV neutrons to $2 \times 10^{22}/\text{m}^2$. The highest E_D was about 0.5 eV, perhaps equivalent to a fast neutron fluence as low as $4 \times 10^{22}/\text{m}^2$ (see Figure 3.2.14 [Söll, 1978]). Other authors would equate this 14-MeV fluence of $2 \times 10^{22}/\text{m}^2$ to a fast neutron fluence of about $11 \times 10^{22}/\text{m}^2$ (Figure 3.4.6(B) [Kulcinski et al., 1986]). However, Snead and Luhman [1986] argued that the high energy neutron irradiation produced less J_c reduction at low fields than the HFBR irradiations for comparable high field reductions. This was interpreted as being due to less resistivity increase per unit E_D for the high energy neutron irradiation owing to the greater Frenkel defect recombination with the more extensive cascade and subcascade structures that occur in 14-MeV irradiation. If recombination is significantly different for 14-MeV and fast neutrons ($E > 0.1$ MeV), fluence conversion is more difficult. A possible example of problems in equating 14-MeV and fast neutron fluence was shown in Figure 2.2.3 for T_c reduction in multifilamentary wire. Another example is the difference in J_c for monofilamentary wire irradiations of different neutron energy shown in Figure 3.4.6(A), although this discrepancy could alternatively be due to a difference in irradiation temperature or a unique material. Without further experimental investigation of conversion, it is not conservative to use 14-MeV results to set radiation limits. In an experimental investigation of such conversions, Snead et al. [1976] measured identical Nb₃Sn samples after both reactor and 14-MeV irradiation and related the changes observed to E_D . They found that E_D was an effective correlation parameter for T_c , but not for I_c .

6.2.2.2. Sensitivity to Magnetic Field and to Magnitude of J_c

Numerous investigations have indicated that J_c is both less enhanced initially and more degraded by irradiation as the field value in which measurements are made is lowered. A theoretical explanation that has been advanced is that

flux pinning forces ($J \times B$) decrease with field so that flux pinning is less effective at lower fields. (The J_c of a type II superconductor is the result of pinning of the lines of magnetic flux that penetrate the material in the mixed state.) Whether or not the theory is correct, considerable experimental evidence supports the increase in degradation with decrease in the magnetic field of measurement, as Figures 3.2.6, 3.2.9, 3.2.10, 3.2.11, 3.3.1, 3.3.2, 3.3.3, and 3.3.4 demonstrate. Therefore, since the 12-T field specified in ITER procurement contracts is lower than the fields in which measurements were made after 14-MeV irradiations of ternary Nb₃Sn [Weiss et al., 1987; Hahn et al., 1991], less initial enhancement and more irradiation degradation should be expected in a 12-T or lower field. On the other hand, the results of cryogenic fast neutron irradiation of ternary Nb₃Sn by Hahn et al. [1986] were obtained in lower fields of 4 and 8 T. Thus, it is conservative to use these results as they stand to assist in setting radiation limits up to 12 T.

The J_c of ternary Nb₃Sn alloys is generally enhanced by increasing ρ_n through the mechanisms expressed in Equations (3.2.1) and (3.2.2) in §3.2.3. However, the higher ρ_n increases the radiation sensitivity of the material, as was shown for the T_c of binary Nb₃Sn by Ghosh et al. [1978] (see Figure 2.1.10). Weiss et al. [1987] and Hahn et al. [1986] attributed the lower fluence for J_c degradation in ternary Nb₃Sn (as compared to binary Nb₃Sn) to the higher initial value of ρ_n that increases with alloying, since the unirradiated wire is already optimized for flux pinning. Possible strain effects were also cited. As noted above, the 15-T values of the prototype ternary Nb₃Sn tested by Hahn et al. [1991] (see Figure 3.4.8) were somewhat below the procurement specifications given in Table 1.3.1. If J_c were increased to meet specifications by a further increase in ρ_n , degradation at a lower radiation fluence would be expected.

6.3. RADIATION LIMITS FROM B_{c2} MEASUREMENTS

Early studies of bulk Nb₃Sn indicated that B_{c2} at 1.6 K fell almost to zero after a fast neutron fluence of $5 \times 10^{23}/\text{m}^2$ ($E > 1$ MeV) [Sweedler et al., 1975] or leveled off to about 17% of the initial value (at 0 K) after a fast neutron fluence of $6 \times 10^{23}/\text{m}^2$ (E not specified) (Figure 4.1.1 [Karkin et al., 1976]). Other ambient-temperature irradiation studies on composite Nb₃Sn

indicated that the decrease in B_{c2} with fluence paralleled that of T_c (Figure 4.2.1 [Ostenson et al., 1983]), with significant degradation of B_{c2} for an in situ composite at $\sim 10^{23}/\text{m}^2$ ($E > 1$ MeV). After a much lower cryogenic fluence of $1.8 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV), B_{c2} had decreased by only about 1 T (Figure 4.2.1 [Colucci and Weinstock, 1978]).

In general, the limited amount of data available on B_{c2} reinforces the conclusions from §§6.1 and 6.2.1 that fast neutron fluences into the 0.5 to $1 \times 10^{23}/\text{m}^2$ range are necessary for significant deterioration of the superconducting properties of binary Nb_3Sn . Although Weiss et al. [1987] reported that enhancements and declines in B_{c2} paralleled those of J_c for both binary and ternary composites (Figures 3.3.3 and 3.3.4), B_{c2} appeared to be a much less significant indicator of ternary radiation damage in the investigation by Hahn et al. [1991]. Figure 3.4.8 shows that J_c values dropped below initial values at a 14-MeV fluence of about $1 \times 10^{22}/\text{m}^2$, and that serious degradation of J_c had occurred at a fluence of $2 \times 10^{22}/\text{m}^2$, yet Table 4.3.1 shows that B_{c2} at this fluence is still high, about 19 T at 4.5 K. This small decrease in B_{c2} is similar to that reported for two of the binary systems tested, although these binary systems exhibited much less deterioration in J_c at the fluence of $2 \times 10^{22}/\text{m}^2$ than the ternary systems. Other measurements of B_{c2} after cryogenic irradiation with fast neutrons are limited to the low fluence data of Colucci and Weinstock [1978] discussed above. With the information available at present, it appears unwise to use B_{c2} measurements to set radiation limits, at least for ternary conductors.

6.4. SYNERGISTIC STRAIN AND RADIATION EFFECTS

Snead and Suenaga [1980] found that T_c of single-core Nb_3Sn decreased more rapidly with neutron irradiation fluence if compressive strain from an adjacent bronze layer was present (Figure 2.5.1).

Irradiation induces changes in the mechanical properties of Nb_3Sn composite constituents that alter the strain dependence of J_c . The strain at failure, ϵ_f , of ternary $(\text{Nb},\text{Ti})_3\text{Sn}$ was found by Katagiri et al. [1993] to decrease to almost half its initial value at a fast neutron fluence of about $3 \times 10^{22}/\text{m}^2$ ($E > 0.1$ MeV). At higher fluences, there was some recovery or saturation in

this trend (Figure 3.5.6). The amount of Ti in this ternary alloy was 1.8 at.%, significantly less than that of the specimens of Hahn et al. [1986], which was 2.6 at.%. In contrast to Figure 3.3.1 from Hahn et al., Figure 3.5.5 shows that the addition of a lower amount of Ti evidently caused less radiation degradation in J_c in comparison to binary specimens. Figure 3.5.5 also indicates that I_c continues to degrade at higher fluences, despite the tendency of ϵ_f to level off after a sharp initial decrease with fluence.

The equations of Summers et al. [1991] on the dependence of T_c , J_c , and B_{c2} on both radiation damage (represented by E_D) and strain (intrinsic and mechanical), have not received a published comparison with experimental data at this writing (§§2.5, 3.5, and 4.3).

6.5. X-RAY STUDIES OF THE ORDER PARAMETER AND LATTICE EXPANSION

Several X-ray studies produced the interesting result that the A15 structure of Nb_3Sn was retained even after irradiation fluences large enough to have displaced each atom from its equilibrium site [Skvortsov et al., 1978; Burbank et al., 1979]. These studies also correlated irradiation damage with antisite defects, the replacement of some Nb atoms on the linear sublattice chain with Sn atoms. Annealing of antisite defects would require high temperatures, ~700 to 900°C, in agreement with numerous experimental observations (e.g., Figures 2.2.2, 2.4.1, 2.4.2, and 2.4.3).

Lattice expansions from 0.5 to 1.25% were found at high fluences [Bauer et al., 1975; Skvortsov et al., 1978].

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